

The composition was prepared in COMPARATIVE EXAMPLE D1 in the same manner as in EXAMPLE D1, except that $\text{C}_6\text{H}_5\text{OSi}(\text{CH}_3)_3$ was not used, and tested in the same manner. The results are given in Table D1.

- 5 The composition was prepared in each of COMPARATIVE EXAMPLES D2 and D3 in the same manner as in EXAMPLE D1, except that $\text{C}_6\text{H}_5\text{OSi}(\text{CH}_3)_3$ was replaced by the same quantity of $(\text{CH}_3)_3\text{SiOH}$ or $(\text{C}_6\text{H}_5)_3\text{SiOH}$, respectively, and tested in the same manner. The results are also given in Table D1.

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Table D1

	Organosilicon compounds	Characteristics						
		H type tensile properties			Tackiness (to the touch)	Storage stability	Curing speed	Resistance to weather
		M ₁₅₀ (kg/cm ²)	T _B (kg/cm ²)	E _B (kg/cm ²)				
EXAMPLE D1	(CH ₃) ₃ SiOC ₆ H ₅	2.6	6.0	720	A	1.01	○	○
COMPARATIVE EXAMPLE D1	Not used	5.3	7.7	320	A	1.25	○	○
COMPARATIVE EXAMPLE D2	(CH ₃) ₃ SiOH	3.0	6.1	460	A	0.97	○	○
COMPARATIVE EXAMPLE D3	(C ₆ H ₅) ₃ SiOH	2.5	5.8	690	B	0.97	○	○

The results given in Table D1 indicate that the composition with $(\text{CH}_3)_3\text{SiOH}$ is lower in improvement extent of modulus and elongation, although excellent in tackiness, and that the composition with $(\text{C}_6\text{H}_5)_3\text{SiOH}$ is deteriorated in tackiness, although improved in modulus and elongation. On the other hand, the composition with $(\text{CH}_3)_3\text{SiOC}_6\text{H}_5$ is improved in modulus and elongation, and, at the same time, excellent in tackiness.

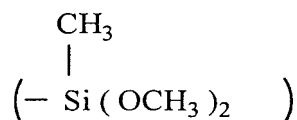
EXAMPLES D2 to D5, and COMPARATIVE EXAMPLES D4 to D6

The composition was prepared in each of the above examples in the same manner as in EXAMPLE D1, except that $\text{C}_6\text{H}_5\text{OSi}(\text{CH}_3)_3$ was replaced by the organosilicon compound shown in Table D2, and tested in the same manner. The results are given in Table D2, together with those of the composition prepared in EXAMPLE D1.

The results given in Table D2 indicate that each composition of the present invention is excellent in all of curing speed, modulus, tackiness, storage stability and resistance to weather, and that use of the organosilicon compound, which generates the compound working as the silanol condensing catalyst, e.g., ammonia or amine, deteriorates storage stability.

REFERENCE EXAMPLE D1

The composition was prepared in the same manner as in EXAMPLE D1, except that 100g of the copolymer rubber (A-1) was replaced by the propylene oxide polymer having, in the molecule, 3 dimethoxysilyl groups on the average, represented by the following formula, and having an average molecular weight of 9,600, and tested in the same manner. The results are given in Table D4.



REFERENCE EXAMPLES D2 to D4

The composition was prepared in REFERENCE EXAMPLE D2 in the same manner as in EXAMPLE D1, except that $\text{C}_6\text{H}_5\text{OSi}(\text{CH}_3)_3$ was not used, and tested in the same manner. The results are given in Table D3.

The composition was prepared in each of REFERENCE EXAMPLES

D3 and D4 in the same manner as in EXAMPLE D1, except that $\text{C}_6\text{H}_5\text{OSi}(\text{CH}_3)_3$ was replaced by the same quantity of $(\text{CH}_3)_3\text{SiOH}$ or $(\text{C}_6\text{H}_5)_3\text{SiOH}$, respectively, and tested in the same manner. The results are also given in Table D3.

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Table D3

	Organosilicon compounds	Characteristics					
		H type tensile properties			Tackiness (to the touch)	Storage stability	Curing speed
		M_{150} (kg/cm ²)	T_B (kg/cm ²)	E_B (kg/cm ²)			
REFERENCE EXAMPLE D1	$(CH_3)_3SiOC_6H_5$	2.5	5.8	710	A	0.92	○ ×
REFERENCE EXAMPLE D2	Not used	5.2	7.6	340	A	1.22	○ ×
REFERENCE EXAMPLE D3	$(CH_3)_3SiOH$	3.5	6.3	490	A	0.92	○ ×
REFERENCE EXAMPLE D4	$(C_6H_5)_3SiOH$	2.5	5.9	680	B	0.92	○ ×

REFERENCE EXAMPLES D5 to D11

The composition was prepared in each of the above examples in the same manner as in REFERENCE EXAMPLE D1, except that $C_6H_5OSi(CH_3)_3$ was replaced by the organosilicon compound shown in Table D4, and tested in the same manner. The results are given in Table D4, together with those of the composition prepared in REFERENCE EXAMPLE D1.

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Table D4

	Organosilicon compounds	Characteristics				
		M ₁₅₀ (kg/cm ²)	Tackiness (to the touch)	Storage stability	Curing speed	Resistance to Weather
REFERENCE EXAMPLE D1	(CH ₃) ₃ SiOC ₆ H ₅	2.5	A	0.92	○	×
REFERENCE EXAMPLE D5	(CH ₃) ₃ SiOCH ₃ Cl	3.3	A	1.02	○	×
REFERENCE EXAMPLE D6	((CH ₃) ₃ SiOCH ₂) ₂	2.2	A	0.92	○	×
REFERENCE EXAMPLE D7	$\begin{array}{c} ((\text{CH}_3)_3\text{SiOCCH}_2\text{CH}_2)_3 \\ \parallel \\ \text{O} \end{array}$	3.3	A	1.18	○	×
REFERENCE EXAMPLE D8	((CH ₃) ₃ SiO) ₃ B	3.1	A	0.92	○	×
REFERENCE EXAMPLE D9	((CH ₃) ₃ Si) ₂ NH	2.7	A	1.38	○	×
REFERENCE EXAMPLE D10	((CH ₃) ₃ Si) ₂ NCH ₃	3.6	A	1.78	○	×
REFERENCE EXAMPLE D11	(CH ₃ SiNH) ₂ CO	4.3	A	1.51	○	×

<EXAMPLES E Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined
5 by the methods described earlier.

Each composition was tested for its curing speed and accelerated weather resistance by the following methods.

(1) Curing speed tests

(1A)

10 Each curing composition was followed for changed frequency by a scanning VNC (SVNC, RAPRA TECHNOLOGY LTD.). Frequency increased with time and became stabilized, and curing speed was based on time required for frequency to change by 95%, wherein the stabilized frequency was set at 100%. The test was conducted
15 at room temperature, in accordance with the instructions described in the following manuals:

(i) Operating manual for the RAPRA's scanning, vibrating probe-type curing tester (scanning VNC) (Software Version 2.2)

(ii) Understanding the RAPRA's scanning, vibrating probe-type
20 curing tester (scanning VNC) (RTL/2844)

(1B)

The curable composition (stock material) was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was 1mm or more and × when it was less than 1mm.

(2) Accelerated weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 under the following conditions, using a Sunshine Carbon Arc weatherometer:

10 <Test conditions>

Light irradiation/rainfall cycles

: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature : $63 \pm 2^\circ\text{C}$

Tank inside temperature : $40 \pm 2^\circ\text{C}$

15 Total light irradiation time : 500 hours

PRODUCTION EXAMPLE E1

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

20 The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied

at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

- 5 The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate
10 the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0-1) thus produced contained ethylene at 68%
15 by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C , iodine value (IV) of 10(g/100g) and M_w/M_n of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the
20 ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the

ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group ($-\text{SiCH}_3(\text{OCH}_3)_2$).

PRODUCTION EXAMPLE E2

5 [Production of silyl-containing vinyl polymer]

Xylene, 340g was charged in a reactor, equipped with an agitator, a thermometer, a nitrogen-supplying nozzle, a drip-feed funnel and a condenser, and heated at 110°C.

10 Then, the reactor was continuously charged for 3 hours with a solution of 140g of styrene, 166g of butyl acrylate, 467g of methyl methacrylate, 100g of stearyl methacrylate, 117g of γ -methacryloxypropyltrimethoxysilane, 10g of N-methylol acrylamide, 30g of γ -mercaptopropyltrimethoxysilane and 30g of azobisisobutyronitrile.

15 On completion of the addition of the monomer, a separately prepared solution of 3g of azobisisobutyronitrile dissolved in 200g of toluene was added to the above mixture in 1 hour for the polymerization for another 1 hour, to produce the silyl-containing vinyl resin.

20 The silyl-containing vinyl resin solution thus prepared contained nonvolatiles at 65%. The resin had a number-average molecular weight of 4,400, determined by GPC.

PRODUCTION EXAMPLE E3

[Production of product of the reaction between an epoxy compound

and an acidic phosphate ester]

Monobutyl phosphate, 50 g (Daihachi Kagaku, MP-4) having an acid value of 670 was charged in a reactor, equipped with an agitator, a thermometer, a nitrogen-supplying nozzle, a drip-feed funnel and a condenser, to which 70.5g of γ -glycidoxypropyltrimethoxysilane was added slowly dropwise in a nitrogen atmosphere, with stirring. After heat generation was no longer observed, the mixture was further heated at 80°C for 1 hour, to continue the reaction. The effluent was incorporated with 12g of methyl orthoacetate, 12g of methanol and 96.5g of xylene, after it was cooled, to produce the curing catalyst containing the active component at 50% (Curing Catalyst 1).

PRODUCTION EXAMPLE E4

[Production of copolymer containing acidic phosphate ester]

Isopropanol, 170g and butyl acetate, 170g were charged in a reactor, equipped with an agitator, a thermometer, a nitrogen-supplying nozzle, a drip-feed funnel and a condenser, and heated at 110°C.

Then, the reactor was continuously charged for 3 hours with a solution of 200g of styrene, 300g of butyl acrylate, 380g of methyl methacrylate, 100g of α -acid phosphooxyethyl methacrylate (Daihachi Kagaku, MR-200), 20g of acrylic acid and 30g of azobisisobutyronitrile.

On completion of the addition of the monomer, a separately prepared solution of 3g of azobisisobutyronitrile dissolved in 200g of butyl acetate was added to the above mixture in 1 hour for the polymerization for another 1 hour. It was further
5 incorporated with 350g of isopropanol, to produce the copolymer containing acidic phosphate ester, containing resin solid at 50% (Curing Catalyst 2).

PRODUCTION EXAMPLE E5

1,9-Decadiene, 138g was charged in a pressure-resistant
10 reactor, to which 256g of trimethoxysilane and 1.04g of 10% isopropanol solution of chloroplatinic acid were added in a nitrogen atmosphere, to allow them to react with each other at 90°C for 4 hours. On completion of the reactions, the product was analyzed by infrared absorption spectroscopy. Infrared
15 absorption of the allyl group at 1640cm^{-1} was found to disappear. The unreacted trimethoxysilane was distilled off at 100°C under a vacuum (5Torr), to obtain the silane compound (B-1) having a structure of $(\text{CH}_3\text{O})_3\text{Si}(\text{CH}_2)_{10}\text{Si}(\text{OCH}_3)_3$.

PRODUCTION EXAMPLE E6

20 1-Octadecene, 252g was charged in a pressure-resistant reactor, to which 142g of trichlorosilane and 0.5g of 10% isopropanol solution of chloroplatinic acid were added in a nitrogen atmosphere, to allow them to react with each other at 90°C for 4 hours. On completion of the reactions, the product

was analyzed by infrared absorption spectroscopy. Infrared absorption of the allyl group at 1640cm^{-1} was found to disappear. The unreacted trichlorosilane was distilled off at 100°C under a vacuum (5Torr). The effluent was incorporated with 192g of

5 methanol, treated under a vacuum to remove hydrogen chloride gas formed, further subjected to ester exchanging at 60°C for 2 hours with the addition of 100g of methyl orthoformate, and treated under a vacuum (5Torr) to distill of the volatiles, to obtain the silane compound (B-2) having a structure of

10 $(\text{CH}_3\text{O})_3\text{Si}(\text{CH}_2)_{17}\text{CH}_3$.

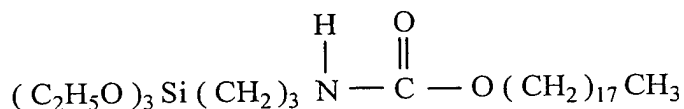
PRODUCTION EXAMPLE E7

1-Octadecanol, 270g was charged in a pressure-resistant reactor, to which 14g of hexane was added, and the mixture was treated for deaeration under a vacuum (5Torr) at 90°C for 1 hour

15 to remove moisture.

It was incorporated with 257g of γ -isocyanate propyltriethoxysilane in a nitrogen atmosphere, and they were allowed to react with each other at 90°C for 2 hours and then at 110°C for 1 hour. The product was analyzed by infrared

20 absorption spectroscopy. Infrared absorption of isocyanate at 2270cm^{-1} disappeared, and that relevant to the urethane bond was observed at 1530cm^{-1} . Thus, it is judged that the silane compound (B-6) having the following structure was formed.

PRODUCTION EXAMPLE E8

5 Hydrogenated polybutadiene glycol, 500g (Nippon Soda Co., Ltd., NISSO-PB GI-1000) having a hydroxyl value of 63.6 was charged in a pressure-resistant reactor, to which 25g of hexane was added, and the mixture was treated for deaeration under a vacuum (5Torr) at 90°C for 1 hour to remove moisture.

10 Next, the above composition was incorporated with 126g of 28% methanol solution of sodium methoxide, and the reactants were allowed to react with each other for 4 hours, while methanol was distilled off at 140°C under a vacuum. Then, 52.5g of allyl chloride was added to the above system dropwise for the reactions

15 at 110°C for 2 hours. The effluent was distilled at 110°C under a vacuum to remove the volatiles. The effluent was incorporated with 1.5L of hexane and 50g of aluminum silicate after it was cooled, and the resultant mixture was stirred for 1 hour, allowed to stand, filtered by celite to remove the salt, and distilled

20 under a vacuum to remove hexane, to obtain the hydrogenated polybutadiene with allyl groups at both terminals.

Then, 300g of the above product was charged in a pressure-resistant reactor, to which 15g of hexane was added, and the mixture was treated for deaeration under a vacuum at

90°C to remove moisture. Next, the above composition was incorporated with 49.9g of trimethoxysilane and 0.21g of 10% isopropanol solution of chloroplatinic acid, to allow them to react with each other at 90°C for 4 hours.

- 5 On completion of the reactions, the effluent was distilled at 100°C under a vacuum to remove the volatiles. This produced the hydrogenated polybutadiene with trimethoxysilyl groups at both terminals (Silane Compound B-3).

EXAMPLES E1 to E7

- 10 The composition containing the dimethoxymethylsilyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE E1, was prepared for each of EXAMPLES E1 to E7. The other components were the
- 15 silane compound (B) having a long polyolefin chain prepared in one of PRODUCTION EXAMPLES E5 to E8, and the curing catalyst (C) prepared in PRODUCTION EXAMPLE E3 or E4, or of a commercial tin compound (dioctyl tin maleate, Sakai Kagaku, TN801TM). The composition for each EXAMPLE is given in Table E1. It was diluted
- 20 with xylene to have an adequate viscosity for a paint.

The coating film of the thermosetting resin was prepared by the following procedure. A soft steel plate was ground by a #240, abrasive paper, coated with a 2-liquid type urethane surfacer (Isamu Toryo, Hiprisurf 2CTM), ground again by a #400

abrasive paper after it was dried, and coated with a clear paint of melamine acrylic resin (Kansai Paint Co., Ltd., Magiclon M-77TM). The coating film was baked at 150°C for 30 minutes, and treated with a medium rubbing compound, to form the base.

5 The coating film of the thermosetting resin was spray-coated with the composition prepared in each of EXAMPLES E1 to E7, after it was made into the paint, by the common method, forcedly dried at 60°C for 30 minutes, and allowed to stand at room temperature for 7 days, to produce the cured coating film.

10 Its adhesion was evaluated by a 2mm square checker pattern cut by a knife, wherein a cellophane tape was put on the pattern and then taken off, for visual observation of the surface exfoliation conditions.

Then, the test piece was placed in a blister box kept at 50°C
15 and RH 98% for 3 days, and evaluated again for adhesion, based on the standards provided by Nippon Paint, Inspection and Testing Association (Point 10: No exfoliation of the coating film, and Point 0: Coating film is totally exfoliated).

The results are given in Table E1.

Table E1

	EXAMPLES						
	E1	E2	E3	E4	E5	E6	E7
Dimethoxysilyl ¹ containing EPDM (1)	100	100	100	100	100	100	100
Silane compound (B) having a long polyolefin chain							
B-1 (PRODUCTION EXAMPLE E5)	6.5	-	-	-	-	-	-
B-2 (PRODUCTION EXAMPLE E6)	-	6.5	-	-	6.5	6.5	6.5
B-3 (PRODUCTION EXAMPLE E7)	-	-	6.5	-	-	-	-
B-4 (PRODUCTION EXAMPLE E8)	-	-	-	6.5	-	-	-
Curing catalyst (C)							
AP-8 *	-	-	-	-	1	-	-
Curing catalyst 1 (PRODUCTION EXAMPLE E3)	2	2	2	2	-	-	-
Curing catalyst 2 (PRODUCTION EXAMPLE E4)	-	-	-	-	-	10	-
TN801 **	-	-	-	-	-	-	2
Adhesion							
Primary	10	10	10	10	10	10	10
Secondary	10	10	10	10	10	10	10
Curing speed (1A) (Hr)	6	7	8	9	7	8	10
(1B)	○	○	○	○	○	○	○
Resistance to weather ***	A	A	A	A	A	A	A

Resistance to weather.	A	A	A
AP-8 (Trade mark), Daihachi Kagaku, A mixture of dioctyl phosphate and monoethyl phosphate			

*** TN801 (Trade mark). Sakai Kagaku, Dioctyl tin maleate

****** TN801 (Trade mark), Sakai Kagaku, Diocetyl tin maleate
******* Evaluation of resistance to weather: A: No cracks or molten portion observed, B: Small cracks or molten portion observed, although slightly,

*** Evaluation of resistance to weathering.
C: Cracks or molten portion observed.

COMPARATIVE EXAMPLES E1 to E11

The composition containing the silyl-containing vinyl resin, prepared in PRODUCTION EXAMPLE E2, was prepared for each of COMPARATIVE EXAMPLES E1 to E11. The other components were the
5 silane compound (B) having a long polyolefin chain prepared in one of PRODUCTION EXAMPLES E5 to E8, and the curing catalyst (C) prepared in PRODUCTION EXAMPLE E3 or E4, or of a commercial tin compound (dioctyl tin maleate, Sakai Kagaku, TN801TM). The composition for each COMPARATIVE EXAMPLE is given in Table E1.
10 It was diluted with xylene to have an adequate viscosity for a paint.

Each composition was evaluated its adhesion in the same manner as in EXAMPLE E1.

The results are given in Table E2.

15 The composition prepared in each of EXAMPLES E1 to E7 and COMPARATIVE EXAMPLES E1 to E11 was tested for curing speed and resistance to weather, in accordance with the methods described earlier.

The results are given in Tables E1 and E2.

Table E2

	COMPARATIVE EXAMPLES										
	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10	E11
Silyl-containing vinyl-based resin (PRODUCTION EXAMPLE E2)	100	100	100	100	100	100	100	100	100	100	100
Silane compound (B) having a long polyolefin chain											
B-1 (PRODUCTION EXAMPLE E5)	6.5	-	-	-	-	-	-	-	-	-	-
B-2 (PRODUCTION EXAMPLE E6)	-	6.5	-	-	6.5	6.5	6.5	-	-	-	-
B-3 (PRODUCTION EXAMPLE E7)	-	-	6.5	-	-	-	-	-	-	-	-
B-4 (PRODUCTION EXAMPLE E8)	-	-	-	6.5	-	-	-	-	-	-	-
Curing catalyst (C)											
AP-8 *	-	-	-	-	1	-	-	1	-	-	-
Curing catalyst 1 (PRODUCTION EXAMPLE E3)	2	2	2	2	-	-	-	-	2	-	-
Curing catalyst 2 (PRODUCTION EXAMPLE E4)	-	-	-	-	-	10	-	-	-	10	-
TN801 **	-	-	-	-	-	-	2	-	-	-	2
Adhesion	10	10	10	10	9	8	8	1	2	0	0
Primary											
Secondary	9	9	9	9	8	7	7	0	0	0	0
Curing speed (Hr) (1A) (1B)	18 x	20 x	24 x	22 x	14 x	16 x	18 x	24 x	36 x	20 x	48 x
Resistance to weather ***	B	B	B	B	C	C	C	C	C	C	C

* AP-8 (Trade mark), Daihachi Kagaku, A mixture of dioctyl phosphate and monoctyl phosphate

** TN801 (Trade mark), Sakai Kagaku, Dioctyl tin maleate

*** Evaluation of resistance to weather: A: No cracks or molten portion observed, B: Small cracks or molten portion observed, although slightly, C: Cracks or molten portion observed.

<EXAMPLES F Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined by the methods described earlier.

The curing speed tests and the weather resistance tests were conducted by the following methods for EXAMPLES and COMPARATIVE EXAMPLES.

(1) Curing speed test

The curable composition (stock material) was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with © when its thickness was 2mm or more, △ when it was 1 to 1.9mm, and × when it was less than 0.9mm.

(2) Weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 using a Sunshine Carbon Arc weatherometer.

<Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

PRODUCTION EXAMPLE F1

[Production of silyl-containing

- 5 ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber ($\text{A}_0\text{-1}$) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random

copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and Mw/Mn of 15.

5 Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the
10 solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group ($-\text{SiCH}_3(\text{OCH}_3)_2$).

15 PRODUCTION EXAMPLE F2

In a 1L metallic autoclave, 148g of phthalic anhydride, 46.4g of propylene oxide, 22.8g of acrylglycidyl ether, 11.6g of allyl alcohol and 0.5g of dimethylbenzylamine were charged and reacted with each other at 100°C for 3 hours, to which 46g of propylene
20 oxide was added for further reactions for another 1 hour. Then, the excess propylene oxide was removed, to obtain the polyester having a molecular weight of 1,200.

Next, 100g of the polyester thus produced was reacted with 9.5g of acetic anhydride at 120°C for 2 hours, and the hydroxyl

group in the polyester was treated, after the excess acetic anhydride was removed. Then 22.2g of the polyester with the treated hydroxyl group was reacted with 0.0035g of chloroplatinic acid and 8.65g of methyl dichlorosilane at 80°C for 3 hours.

5 The excess methyl dichlorosilane was removed under a vacuum. Then, the above effluent was incorporated with 20mL of methanol and 20mL of methyl orthoformate, and the mixture was stirred at room temperature for 1 hour to remove the low-boiling materials under a vacuum. This produced the silyl-containing polyester.

10 PRODUCTION EXAMPLE F3

A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 16g of allyl methacrylate, 20g of methyl methacrylate, 19g of n-butyl methacrylate, 14g of n-butyl acrylate, 1g of acrylic acid and 2g of n-dodecylmercaptan was added dropwise
15 to 100g of toluene as the solvent heated at 90°C, and they were allowed to react with each other for 10 hours, to obtain the vinyl-based polymer having a molecular weight of 8,000 and containing an allyl type unsaturated group.

The vinyl-based polymer had the infrared absorption relevant
20 to the carbon-carbon double bond at 1648cm^{-1} .

A solution of 1.5g of methyldimethoxysilane and 0.0005g of chloroplatinic acid dissolved in isopropanol was added to 20g of the vinyl-based polymer having an allyl type unsaturated group thus produced, and they were allowed to react with each other

at 90°C for 6 hours under sealed conditions. The product had no infrared absorption at 1648cm^{-1} in the infrared absorption spectral pattern. Therefore, it was judged that the silyl-containing vinyl-based polymer was produced.

5 PRODUCTION EXAMPLE F4

The silyl-containing diallyl phthalate-based copolymer was produced in the same manner as in PRODUCTION EXAMPLE F3, except that 16g of allyl methacrylate was replaced by 31g of diallyl phthalate.

10 PRODUCTION EXAMPLE F5

A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 27g of γ -methacryloxypropyltrimethoxysilane, 20g of methyl methacrylate, 19g of n-butyl methacrylate, 14g of n-butyl acrylate, 1g of acrylic acid and 2g of n-dodecylmercaptan was added dropwise to 100g of toluene as the solvent heated at 100°C, and they were allowed to react with each other for 10 hours, to obtain the silyl-containing vinyl-based polymer having a molecular weight of 9,000.

PRODUCTION EXAMPLE F6

20 One hundred grams of diallyl phthalate prepolymer (DAISO Co. Ltd., DAISO DAP LTM) having an iodine value of around 80, 0.00001g of chloroplatinic acid and 1g of hydroquinone were dissolved in 100mL of toluene, to which 35mL of methyl diethoxysilane was added, and they were allowed to react with each other at 90°C

for 3 hours, to obtain the silyl-containing diallyl phthalate prepolymer.

PRODUCTION EXAMPLE F7

[Production of silyl-containing polymer]

5 Xylene, 45.9 parts by weight was charged in a reactor, equipped with an agitator, a thermometer, a reflux condenser, an N₂ gas-supplying nozzle and a drip-feed funnel, and heated to 110°C in a flow of N₂ gas, to which the mixture (a) described below was added dropwise through the drip-feed funnel at a constant rate for 5 hours:

Mixture (a);

Styrene	12.8 parts by weight
Methyl methacrylate	50.1 parts by weight
Stearyl methacrylate	6.9 parts by weight
15 γ -Methacryloxypropyltrimethoxysilane	30.2 parts by weight

Xylene	13.5 parts by weight
--------	----------------------

2,2'-Azobisisobutyronitrile	4.5 parts by weight
-----------------------------	---------------------

On completion of addition of the above mixture (a), 0.5 part by weight of 2,2'-azobisisobutyronitrile and 5 parts by weight of toluene were further added at a constant rate for 1 hour. The resultant resin solution was cured at 110°C for 2 hours and cooled, to which xylene was added to adjust the solid content at 60%.

The characteristics of the Resin Solution A thus obtained are given in Table F1.

PRODUCTION EXAMPLE F8

[Production of acrylic-based resin for paints]

5 Resin Solution B was produced in the same manner as in PRODUCTION EXAMPLE F7, except that 31.8 parts by weight of butyl acetate and 9.5 parts by weight of xylene were charged, to which the mixture (b) described below was added in place of the mixture (a):

10 Mixture (b)

Xylene	18.0 parts by weight
Styrene	28.3 parts by weight
Methyl methacrylate	6.9 parts by weight
n-Butyl acrylate	47.6 parts by weight
15 Methacrylic acid	0.3 part by weight
2-Hydroxyethyl methacrylate	16.9 parts by weight
2,2'-Azobisisobutyronitrile	1.8 parts by weight

On completion of addition of the above mixture (b), 0.2 part by weight of 2,2'-azobisisobutyronitrile and 3.8 parts by weight
20 of toluene were further added at a constant rate for 1 hour. The resultant resin solution was cured at 110°C for 2 hours and cooled, to which xylene was added to adjust the solid content at 60%.

The characteristics of the Resin Solution B thus obtained

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are given in Table F1.

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Table F1

	Resin Solution A	Resin Solution B
Characteristics		
Non-volatile matters [%]	60	60
Viscosity (23°C) [cPs]	900	4400
Acid value (KOH/g solid)	0	2.0
Hydroxyl value (KOH/g solid)	0	73
Color Number (Gardner)	< 1	< 1

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EXAMPLES F1 to F8, and COMPARATIVE EXAMPLES F1 to F14

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A soft steel plate as the base for the test piece was degreased, ground by a #240 abrasive paper, coated with a urethane surfacer, and baked at 80°C for 30 minutes. Then, the coated surface was further ground by a #600 abrasive paper, coated with a clear paint, described in Table F2 for each of EXAMPLES F1 to F8 and COMPARATIVE EXAMPLES F1 to F14, and baked at 140°C for 30 minutes, to prepare the test piece.

Each test piece was left at room temperature for 30 minutes, after it was baked under the above conditions. Their characteristics are given in Table F2.

The notes *1 to *4 in Table F2 are described below:

*1 : Q-631 is a modified cycloaliphatic polyamine, produced by Mitsui Chemicals, Inc.

*2 : Hardness was determined in accordance with JIS K-5400.

*3 : For the toluene spot test, several drops of toluene were dropped onto the coating film, left at room temperature and dried, to observe the coating film conditions.

Resistance of the film to the solvent was evaluated according to the following four-grade system.

<Four-grade system for evaluation of the resistance to the solvent>

◎ : No change is observed at all on the coating film surface

○ : No change is observed on the coating film surface

△ : Traces are left on the coating film surface

× : The coating film is dissolved

*4 : Evaluation of resistance to weather

5 A: No cracks or molten portion observed.

B: Small cracks or molten portion observed, although slightly.

C: Cracks or molten portion observed.

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Table F2 (1)

	EXAMPLES							
	F1	F2	F3	F4	F5	F6	F7	F8
Composition (parts by weight) Silyl-Containing Copolymer PRODUCTION EXAMPLE F1	100	100	100	100	100	100	100	100
Acrylic Resin PRODUCTION EXAMPLE F8	80	-	-	-	-	-	-	-
Thermosetting Acrylic Paint (Belcoat No.5200)	-	80	-	-	-	80	-	80
Alkyd paint (Hariphthal SFC42-60X)	-	-	40	-	-	-	-	-
Epoxy-based paint (Epikote 1001)	-	-	-	40	-	-	-	-
Organopolysiloxane (Z6018)	-	-	-	-	40	-	40	-
Amines(B) Q-631 * 1	-	-	-	-	-	2	2	-
Piperidine	-	-	-	-	-	-	-	3
Monoethanol amine	2	2	2	2	2	-	-	-
Silane-coupling agent (C) N-β-(aminoethyl)-γ aminopropyl trimethoxysilane	-	-	-	-	-	1	1	-
γ-aminopropyltriethoxysilane	-	-	-	-	-	-	-	1.5
γ-mercaptopropyltriethoxysilane	1	1	1	1	1	-	-	-
Tests								
Hardness * 2	3H	3H	3H	3H	3H	3H	3H	3H
Toluene spot test * 3	○ to ⊙	○ to ⊙	○ to ⊙	○ to ⊙	○ to ⊙	○ to ⊙	○ to ⊙	○ to ⊙
Curing speed (Film tension)	⊙	⊙	⊙	⊙	⊙	⊙	⊙	⊙
Resistance to weather * 4	A	A	A	A	A	A	A	A

Table F2 (2)

	COMPARATIVE EXAMPLES						
	F1	F2	F3	F4	F5	F6	F7
Composition (parts by weight) Silyl-Containing Copolymer PRODUCTION EXAMPLE F3	-	-	-	-	-	100	100
PRODUCTION EXAMPLE F4	-	-	-	-	-	-	-
PRODUCTION EXAMPLE F5	100	100	100	100	100	-	-
Acrylic Resin PRODUCTION EXAMPLE F8	80	-	-	-	-	-	-
Thermosetting acrylic paint (Belcoat No.5200)	-	80	-	-	-	80	-
Alkyd paint (Hariphthal SFC42-60X)	-	-	40	-	-	-	-
Epoxy-based paint (Epikote 1001)	-	-	-	40	-	-	40
Organopolysiloxane (Z6018)	-	-	-	-	40	-	-
Amines (B) Q-631 * 1	-	-	-	-	-	2	2
Piperidine	-	-	-	-	-	-	-
Monoethanolamine	2	2	2	2	2	-	-
Silane coupling agent (C) N- β -(aminoethyl)- γ -aminopropyl- Trimethoxysilane	-	-	-	-	-	1	1
γ -Aminopropyltriethoxysilane	-	-	-	-	-	-	-
γ -Mercaptopropyltriethoxysilane	1	1	1	1	1	-	-
Tests							
Hardness * 2	2H	2H	2H	2H	2H	2H	2H
Toluene spot test * 3	O	O	O	O	O	O	O
Curing speed (Film tension)	x	x	x	x	x	x	x
Resistance to weather * 4	B	B	B	B	B	B	B

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Table F2 (3)

	COMPARATIVE EXAMPLES							
	F8	F9	F10	F11	F12	F13	F14	
Composition (parts by weight) Silyl-Containing Copolymer								
PRODUCTION EXAMPLE F2	-	-	-	-	-	-	100	
PRODUCTION EXAMPLE F3	-	-	-	-	-	-	-	
PRODUCTION EXAMPLE F4	100	-	-	-	-	-	-	
PRODUCTION EXAMPLE F5	-	100	100	-	-	-	-	
Acrylic Resin	-	-	-	-	-	-	80	
Thermosetting acrylic paint (Belcoat No.5200)	80	-	80	180	-	-	-	
Alkyd paint (Hariphthal SFC42-60X)	-	-	-	-	-	-	-	
Epoxy-based paint (Epikote 1001)	-	-	-	-	140	-	-	
Organopolysiloxane (Z6018)	-	-	-	-	-	140	-	
Amines (B)	-	-	-	-	-	-	-	
Q-631 * 1	-	-	-	-	-	-	-	
Piperidine	3	-	-	-	-	-	-	
Monoethanolamine	-	2	-	2	2	2	2	
Silane-coupling agent (C)	-	-	-	-	-	-	-	
N-β-(aminoethyl)-γ aminopropyl trimethoxysilane	-	-	-	-	-	-	-	
γ-aminopropyltriethoxysilane	1.5	-	-	-	-	-	-	
γ-mercaptopropyltriethoxysilane	-	1	-	1	1	1	1	
Tests								
Hardness * 2	2H	HB	6B	2H	2H	2H	3H	
Toluene spot test * 3	○	△	×	○	○	○	○ to ⊙	
Curing speed (Film tension)	×	×	×	×	×	×	×	
Resistance to weather * 4	B	C	C	B to C	B to C	B to C	C	

REFERENCE EXAMPLES F1 to F5, and REFERENCE COMPARATIVE EXAMPLES
F1 to F5

A soft steel plate as the base for the test piece was degreased, ground by a #240 abrasive paper, coated with a urethane surfacer, and baked at 80°C for 30 minutes. Then, the coated surface was further ground by a #600 abrasive paper, coated with a clear paint, described in Table F3 for each of REFERENCE EXAMPLES F1 to F5 and REFERENCE COMPARATIVE EXAMPLES F1 to F5, and baked at 140°C for 30 minutes, to prepare the test piece.

Each test piece was left at room temperature for 30 minutes, after it was baked under the above conditions. Their characteristics are given in Table F3.

The notes *1 to *11 in Table F3 are described below:

- *1 : Thermosetting acrylic paint, produced by NOF Corp.
- *2 : Soybean fatty acid short-oil type alkyd resin, produced by Harima Chemicals.
- *3 : Epoxy resin, produced by Shell
- *4 : Organopolysiloxane, produced by Dow Corning
- *5 : Dioctyl tin maleate, Sakai Kagaku Kogyo
- *6 : Amino-containing silane coupling agent, produced by UCC
- *7 : Epoxy-containing silane coupling agent, produced by UCC
- *8 : Dioctyl acid phosphate, produced by Daihachi Kagaku
- *9 : Hardness was determined in accordance with JIS K-5400
- *10 : For the toluene spot test, several drops of toluene was

dropped onto the coating film, left at room temperature and dried, to observe the coating film conditions. Resistance of the film to the solvent was evaluated according to the following four-grade system.

5 <Four-grade system for evaluation of the resistance to the solvent>

◎ : No change is observed at all on the coating film surface

○ : No change is observed on the coating film surface

10 △ : Traces are left on the coating film surface

× : The coating film is dissolved

*11 : Evaluation of resistance to weather

A: No cracks or molten portion observed

15 B: Small cracks or molten portion observed, although slightly

C: Cracks or molten portion observed

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	REFERENCE EXAMPLE				
	F1	F2	F3	F4	F5
Composition (parts by weight) Silyl-Containing Copolymer PRODUCTION EXAMPLE F7	100	100	100	100	100
Acrylic Resin PRODUCTION EXAMPLE F8 Thermosetting acrylic paint (Belcoat No.5200, Clear S) *1	80	-	-	-	-
Alkyd paint (Hariphthal SFC42-60X) *2	-	-	40	-	-
Epoxy-based paint (Epikote 1001) *3	-	-	-	40	-
Organopolysiloxane (Z6018) *4	-	-	-	-	40
Curing catalysts TN801 *5	4	4	-	-	-
A-1120 *6	1	1	-	-	-
A-187 *7	1	1	-	-	-
DP-8 *8	-	-	1	1	1
N,N'-dimethyl-n-dodecylamine	-	-	1	1	1
Tests					
Hardness *9	2H	2H	2H	2H	2H
Toluene spot test *10	O	O	O	O	O
Curing speed (Film tension)	x	x	x	x	x
Resistance to weather *11	B	B	B	B	B

Table F3 (2)

	REFERENCE COMPARATIVE EXAMPLE				
	F1	F2	F3	F4	F5
Composition (parts by weight) Silyl-Containing Copolymer PRODUCTION EXAMPLE F7	100	100	-	-	-
Acrylic Resin PRODUCTION EXAMPLE F8	-	80	-	-	-
Thermosetting acrylic paint (Belcoat No.5200, Clear S) *1	-	-	180	-	-
Alkyd paint (Hariphthal SFC42-60X) *2	-	-	-	-	-
Epoxy-based paint (Epikote 1001) *3	-	-	-	140	-
Organopolysiloxane (Z6018) *4	-	-	-	-	140
Curing catalysts	4	-	4	-	-
TN801 *5	1	-	1	-	-
A-1120 *6	1	-	1	-	-
A-187 *7	-	-	-	1	1
DP-8 *8	-	-	-	1	1
N,N-dimethyl n-dodecylamine	-	-	-	1	1
Tests	HB	6B	2H	2H	2H
Hardness *9	Δ	×	○	○	○
Toluene spot test *10	×	×	×	×	×
Curing speed (Film tension)	C	C	C	C	C
Resistance to weather *11					

<EXAMPLES G Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (M_w/M_n) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined
5 by the methods described earlier.

PRODUCTION EXAMPLE

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
rubber (A-1)]

10 The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
propylene and 5-vinyl-2-norbornene were continuously supplied
15 at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $VO(OEt)_2Cl$
and $Al(Et)_{1.5}Cl_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
per hour, respectively, also continuously.

The copolymerization effected under the above conditions
20 produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
the polymerization. The polymer was separated from the solvent

by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68%
5 by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and Mw/Mn of 15.

Two% toluene solution (0.3 part by weight) of chloroplatinic acid and 1.5 parts by weight of methyldimethoxysilane were added
10 to 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This
15 produced 101.5 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group ($-\text{Si}(\text{CH}_3)(\text{OCH}_3)_2$).

EXAMPLES G1 to G8, and REFERENCE EXAMPLE G1

20 A mixture containing the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE, was prepared for each of the above examples. It was composed of 100g of the copolymer rubber (A-1), 120g of calcium carbonate as the filler

(Shiraishi K.K., CCRTM), 20g of titanium dioxide (Ishihara Sangyo Kaisha, Ltd., R820TM), 2g of dibutyl tin diacetylacetonate (NITTO KASEI, U-220TM) as the curing promoter, 50g of a paraffin-based process oil as the plasticizer (Idemitsu Kosan, Diana Process Oil PW-380TM) for EXAMPLES G1 to G8 and REFERENCE EXAMPLE G1, 2g of a monovalent silanol compound shown in Table G1, and N-(β -aminoethyl)- γ -aminopropyltrimethoxy silane ($\text{H}_2\text{NCH}_2\text{CH}_2\text{NHCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$) and N-(β -aminoethyl)- γ -aminopropylmethyldimethoxysilane ($\text{NH}_2\text{CH}_2\text{CH}_2\text{NHCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{CH}_3)(\text{OCH}_3)_2$) as the trifunctional and bifunctional aminosilane compound, respectively, in a quantity shown in Table G1. It was sufficiently kneaded to mix the components by a 3-paint roll unit, and put in the H-shape test piece prepared in accordance with JIS A-5758, to determine its tensile characteristics, adhesion strength and resistance of the adhesion strength to weather while the test piece was irradiated with light (weather-resistant adhesion). It was also analyzed for its curing speed and resistance to weather (ozone-caused aging test). The results are given in Table G1.

The following testing methods were used.

(Tensile characteristics)

The composition thus prepared was put in the H-shape test piece prepared in accordance with JIS A-5758 (base: anodized aluminum oxide), which was cured at 23°C and RH60% for 14 days,

and further cured at 30°C for 14 days, to be tensile-tested at a speed of 30mm/minute.

(Adhesion strength)

The test piece, when fractured in the tensile test, was observed
5 for the fractured conditions.

It is judged to have a high adhesion strength to the base, when the cured product itself was fractured (cohesion fracture, CF), and a low adhesion strength when the cured product and base were separated from each other at the adhesion interface (adhesion
10 fracture, AF).

(Weather-resistant adhesion)

The H-shape test piece (base: glass) was prepared in accordance with JIS A-5758, irradiated with light for 480 hours for accelerated exposure test by a Sunshine weatherometer (Suga
15 Shikenki, WEL-3-HC), and tensile-tested by an Autograph (Shimadzu, IS-5000).

(Curing speed test)

The curable composition was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

20 Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was more than 1mm, △ when it was 0.5 to 1mm, and × when it was less than 0.5mm.

(Weather resistance test)

The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions.

Analyzer : Sunshine Carbon Arc weatherometer

5 Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^{\circ}\text{C}$

Tank inside temperature: $40 \pm 2^{\circ}\text{C}$

Total light irradiation time: 500 hours

10 The tested test piece was visually observed, to evaluate its resistance to weather according to the following four grades:

◎ : No cracks or molten portion observed

○ : Cracks or molten portion observed, although slightly

△ : Cracks or molten portion observed to some extent

15 × : Cracks or molten portion observed significantly

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Table G1

	Monovalent silanol-based compound	Trifunctional Aminosilane (g)		Hf type tensile characteristics			Adhesive strength to aluminum	Weather-resistant adhesion			Curing speed	Resistance to weather
		Bifunctional Aminosilane (g)		M ₅₀ (kg/cm ²)	T _B (kg/cm ²)	E _H (%)		T _B (kg/cm ²)	E _B (%)	Adhesion conditions		
EXAMPLE G1	$\text{CH}_3\text{C} \begin{array}{c} \diagup \text{OSi}(\text{CH}_3)_3 \\ \diagdown \text{NSi}(\text{CH}_3)_3 \end{array}$	0.5 — 3.0		2.0	8.9	550	CF	8.7	530	CF	○	◎
EXAMPLE G2	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	1.0 — 3.0		2.4	8.5	530	CF	8.3	510	CF	○	◎
EXAMPLE G3	$(\text{CH}_3)_3\text{Si-O-C}_6\text{H}_5$	0.5 — 3.0		2.6	8.4	560	CF	8.2	530	CF	○	◎
EXAMPLE G4	-	0.5 — 3.0		3.9	8.6	520	CF	8.2	500	CF	△	○
EXAMPLE G5	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	3.0 — 0		5.8	9.4	320	CF	8.9	290	CF	○	○
EXAMPLE G6	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	0 — 3.0		2.8	8.2	450	CF	7.9	400	AF	○	○
REFERENCE EXAMPLE G1	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	0 — 0		4.9	-	-	AF	-	-	AF	○	○
EXAMPLE G7	-	0 — 3.0		5.1	-	-	AF	-	-	AF	△	○
EXAMPLE G8	-	3.0 — 0		6.2	-	-	AF	-	-	AF	△	○

In the table, the bar "--" was for a test piece with the composition separated from the base and the properties of the cured product itself were immeasurable.

The comprehensive evaluation results of the compositions prepared in EXAMPLES G1 to G8 and REFERENCE EXAMPLE G1 are given in Table G2, wherein those marked with ○ have good characteristics, × have not and △ are in-between.

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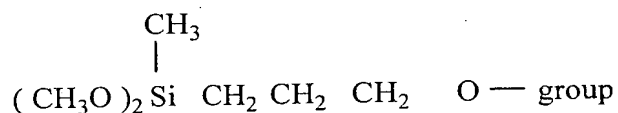
Table G2

	Monovalent silanol-based compound	Trifunctional aminosilane(g)		Modulus	Adhesive strength	Weather-resistant adhesion	Curing speed	Resistance to weather
		Bifunctional aminosilane(g)						
EXAMPLE G1	Used	0.5/3.0		○	○	○	○	○
EXAMPLE G2	Used	1.0/3.0		○	○	○	○	○
EXAMPLE G3	Used	0.5/3.0		○	○	○	○	○
EXAMPLE G4	Not used	0.5/3.0		△	○	○	△	△
EXAMPLE G5	Used	3.0/0		×	○	○	△	△
REFERENCE EXAMPLE G1	Used	0/3.0		○	○	×	△	△
EXAMPLE G6	Used	0/0		×	×	×	△	△
EXAMPLE G7	Not used	0/3.0		×	×	×	△	△
EXAMPLE G8	Not used	3.0/0		×	×	×	△	△

COMPARATIVE PRODUCTION EXAMPLE

Eight hundred g of a polyoxypropylene-based polymer, with allyl ether group at 97% of the total terminals and having an average molecular weight of around 8,000, was charged in a pressure-resistant reactor equipped with an agitator, to which 19g of methyldimethoxysilane was added. The mixture was then incorporated with 0.34mL of a solution of chloroplatinic acid ($\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$), 8.9g dissolved in 18mL of isopropyl alcohol and 160mL of tetrahydrofuran, and they were allowed to react with each other at 80°C for 6 hours.

The quantitative analysis by IR spectroscopy indicated that the hydrogenated silicon group little remained in the reaction solution. The quantitative analysis of the reactive silicon group by NMR indicated that the polyoxypropylene-based polymer (CA-1) produced had approximately 1.7 groups represented by the following formula on the average in one molecule at the terminal.

REFERENCE EXAMPLES G2 to G10

The curable composition was prepared for each of REFERENCE EXAMPLES G2 to G10 in the same manner as in EXAMPLES G1 to G8 and REFERENCE EXAMPLE G1, except that the silyl-containing

ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) was replaced by the polymer (CA-1) prepared in COMPARATIVE PRODUCTION EXAMPLE, and the paraffin-based process oil as the plasticizer (Idemitsu Kosan, Diana Process Oil PW-380TM) was replaced by the oxypropylene polymer of the allyl ether type at the terminal, having an Mn of 5,200 and Mw/Mn of 1.6 for REFERENCE EXAMPLES G2 to G3 and G5 to G10), or (2-ethylhexyl) phthalate (Daihachi Kagaku) for REFERENCE EXAMPLE G4. The characteristics of these compositions were evaluated. The results are given in Table G3.

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Table G3

	Monovalent silanol-based compound	Trifunctional aminosilane (g) Bifunctional aminosilane (g)	H type tensile characteristics			Adhesive strength to aluminum	Weather-resistant adhesion			Curing speed	Resistance to weather
			M ₁₅₀ (kg/cm ²)	T _B (kg/cm ²)	E _B (%)		T _B (kg/cm ²)	E _B (%)	Adhesion conditions		
REFERENCE EXAMPLE G2	$\text{CH}_3\text{C} \begin{array}{c} \text{OSi}(\text{CH}_3)_3 \\ \diagdown \\ \text{NSi}(\text{CH}_3)_3 \end{array}$	0.1 — 2.0	2.9	8.3	460	CF	8.0	420	CF	○	△ to ×
REFERENCE EXAMPLE G3	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	0.5 — 2.0	2.9	7.9	420	CF	7.7	400	CF	○	△ to ×
REFERENCE EXAMPLE G4	$(\text{CH}_3)_3\text{Si-O-C}_6\text{H}_5$	0.5 — 2.0	2.8	7.8	440	CF	7.2	380	CF	○	△ to ×
REFERENCE EXAMPLE G5	—	0.5 — 2.0	3.9	8.2	460	CF	8.1	440	CF	△	×
REFERENCE EXAMPLE G6	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	2.0 — 0	5.3	9.1	330	CF	8.9	300	CF	○	×
REFERENCE EXAMPLE G7	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	0 — 2.0	2.8	8.1	440	CF	(1.9)	(120)	AF	○	×
REFERENCE EXAMPLE G8	$(\text{CH}_3)_3\text{Si-NH-Si}(\text{CH}_3)_3$	0 — 0	—	(2.0)	(90)	AF	(0.9)	(60)	AF	○	×
REFERENCE EXAMPLE G9	—	0 — 2.0	4.7	9.2	400	CF	(1.5)	(110)	AF	△	×
REFERENCE EXAMPLE G10	—	2.0 — 0	5.8	9.6	320	CF	8.1	290	CF	△	×

In the table, the value in the parentheses () was for a test piece with the composition separated from the base at the adhesion interface, and consequently the properties are not of the cured product itself.

- 5 The comprehensive evaluation results of the compositions prepared in EXAMPLES G2 to G10 are given in Table G4, wherein those marked with ○ have good characteristics, × have not and △ are in-between.

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Table G4

		Monovalent silanol-based compound	Trifunctional Aminosilane (g)		Modulus	Adhesive strength	Weather-resistant adhesion	Curing speed	Resistance to weather
			Bifunctional Aminosilane (g)						
REFERENCE EXAMPLE G	2	Used	0.1/2.0		○	○	○	×	×
	3	Used	0.5/2.0		○	○	○	×	×
	4	Used	0.5/2.0		○	○	○	×	×
	5	Not used	0.5/2.0		×	○	○	×	×
	6	Used	2.0/0		×	○	○	×	×
	7	Used	0/2.0		○	○	×	×	×
	8	Used	0/0		-	×	×	×	×
	9	Not used	0/2.0		×	○	×	×	×
	10	Not used	2.0/0		×	○	○	×	×

<EXAMPLES H Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLE and COMPARATIVE EXAMPLE were determined
5 by the methods described earlier.

PRODUCTION EXAMPLE

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
rubber (A-1)]

10 The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied
15 at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions
20 produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent

by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68%
5 by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and Mw/Mn of 15.

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10 Two% toluene solution (0.3 part by weight) of chloroplatinic acid and 1.5 parts by weight of methyldimethoxysilane were added to 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This
15 produced 101.5 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group ($-\text{Si}(\text{CH}_3)(\text{OCH}_3)_2$).

EXAMPLE H1 and COMPARATIVE EXAMPLE H1

20 The one-liquid type curable composition was prepared using the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) prepared in PRODUCTION EXAMPLE, to evaluate its storage stability and resistance of the cured product to weather.

Their compositions are given in Table H1 (parts by weight), and the evaluation results in Table H2.

It was evaluated by the following methods.

(Viscosity and curing speed (tack-free time))

5 These were determined in accordance with JIS A-5758.

(Curing speed test)

The curable composition was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

10 Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was 1mm or more and × when it was less than 1.0mm.

(Weather resistance test)

15 The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions.

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

20 Black panel temperature: 63±2°C

Tank inside temperature: 40±2°C

Total light irradiation time: 500 hours

The tested test piece was visually observed, to evaluate its resistance to weather according to the following two-grade

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system: ○: No cracks or molten portion observed, and ×: Cracks or molten portion observed.

(Storage stability)

Storage stability of the curable composition was evaluated
5 by viscosity, i.e., ratio of viscosity of the composition stored at 50°C for 4 weeks in a nitrogen-purged container to that of the one immediately after its was prepared.

The composition having the ratio closer to unity (1) means it is more excellent in storage stability.

10 REFERENCE EXAMPLES H1 to H3

15 The one-liquid type curable composition was prepared for each of REFERENCE EXAMPLES H1 to H3 using the oxypropylene polymer having around 2 dimethoxymethylsilyl groups ($-\text{Si}(\text{CH}_3)(\text{OCH}_3)_2$) in the molecule and an average molecular weight of 9,000 (KANEKA CORP., MS Polymer, hereinafter referred to as CA-1) and
oxypropylene polymer having around 1.5 dimethoxysilyl groups in the molecule and an average molecular weight of 8,000 (KANEKA CORP., MS Polymer, hereinafter referred to as CA-2), and
evaluated in the same manner as in EXAMPLE H1. Their compositions
20 are given in Table H1 (parts by weight), and the evaluation results in Table H2.

Table H1

		EXAMPLE	COMPARATIVE EXAMPLE	REFERENCE EXAMPLE	REFERENCE EXAMPLE	REFERENCE EXAMPLE
	Components	H1	H1	H1	H2	H3
Polymer rubber	A-1	100	100	0	0	0
	CA-1	0	0	50	50	50
	CA-2	0	0	50	50	50
Filler	Colloidal calcium Carbonate *1	120	120	120	120	120
	Titanium oxide *2	20	20	20	20	20
Plasticizer	Paraffin-based oil* 3	50	50	50	50	50
Dehydrator	VTMO* 4	3	3	3	3	3
Tackifier	DAMO* 5	2	2	2	2	2
Curing catalyst	U-220* 6	1	1	1	1	1
Organic carboxylic acid	2-Ethylhexanoic acid	0.2	0	0.2	0	0
	Stearic acid	0	0	0	0.4	0

*1 CALFORT-S (STURGE)

*2 TIOFWE R85 (TDF)

*3 Paraffin-based oil (Idemitsu Kosan, Diana Process Oil PW-3™)

*4 DYNASYLAN VTMO (Huls)

*5 DYNASYLAN DAMO (Huls)

*6 Dibutyl tin diacetylacetate (NITTO KAGAKU)

Table H2

	Characteristics	EXAMPLE H1	COMPARATIVE EXAMPLE H1	REFERENCE EXAMPLE H1	REFERENCE EXAMPLE H2	REFERENCE EXAMPLE H3
Immediately after the test piece was prepared	Viscosity (Poise)	12000	11000	13000	11000	9500
	Tack-free time (hr)	4	2.5	5.5	4.5	2.5
	Curing speed	○	○	○	○	○
	Resistance to weather	○	○	×	×	×
After the test piece was stored at 50°C for four weeks	Viscosity (Poise)	14000	15000	13000	13000	13000
	Tack-free time (hr)	4	8	5.5	5.0	>10
	Curing speed	○	○	○	○	×
	Resistance to weather	○	○	×	×	×
Storage stability	Viscosity ratio	1.17	1.36	1.18	1.18	1.37

<EXAMPLES J Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLE and EXAMPLE were determined by the methods described earlier.

The curing speed tests and accelerated weather resistance tests were conducted by the following methods for EXAMPLES and REFERENCE EXAMPLES.

(1) Curing speed test

The curable composition (stock material) was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was 1mm or more, and × when it was less than 1mm.

(2) Accelerated weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 using a Sunshine Carbon Arc weatherometer.

<Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

Property measurements: in accordance with JIS K-6301

PRODUCTION EXAMPLE J1

- 5 [Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
rubber (A-1)]

10 The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
propylene and 5-vinyl-2-norbornene were continuously supplied
at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$
15 and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
per hour, respectively, also continuously.

The copolymerization effected under the above conditions
produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber (A₀-1) in a form of uniform solution.

- 20 A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
the polymerization. The polymer was separated from the solvent
by steam-stripping the solution, and dried at 55°C for 48 hours
under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and
5 Mw/Mn of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other
10 at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group
15 (-SiCH₃(OCH₃)₂).

PRODUCTION EXAMPLE J2

Methyl methacrylate, 0.2mol, allyl methacrylate, 0.086mol and n-dodecyl mercaptan, 5g were dissolved in 70mL of toluene, to which 0.5g of azobisisobutyronitrile was added, and they were
20 allowed to react with each other at 80°C for 4 hours (the toluene solution could be directly used for the subsequent hydrosilylation). The solvent was distilled off under a vacuum, in order to obtain the acrylic-based polymer having a molecular weight of around 2,000 and containing an allyl type unsaturated

group.

The far-infrared absorption spectroscopic analysis indicated that the acrylic-based polymer thus produced had the absorption relevant to the carbon-carbon double bond at 1648cm^{-1} , in addition to the strong absorption relevant to the ester at 1730cm^{-1} .

PRODUCTION EXAMPLE J3

Methyl methacrylate, 0.2mol, allyl acrylate, 0.086mol and n-dodecyl mercaptan, 5g were dissolved in 70mL of toluene, to which 0.5g of azobisisobutyronitrile was added, and they were allowed to react with each other at 80°C for 4 hours. This produced the acrylic-based polymer having a molecular weight of around 2,000 and containing an allyl type unsaturated group.

The far-infrared absorption spectroscopic analysis indicated that the acrylic-based polymer thus produced had the absorption relevant to the carbon-carbon double bond at 1648cm^{-1} .

PRODUCTION EXAMPLE J4

Methyl methacrylate, 0.1mol, styrene, 0.1mol, allyl methacrylate, 0.086mol and n-dodecyl mercaptan, 5g were dissolved in 70mL of toluene, to which 0.5g of azobisisobutyronitrile was added, and they were allowed to react with each other at 80°C for 4 hours. This produced the vinyl copolymer having a molecular weight of around 2,000.

The far-infrared absorption spectroscopic analysis indicated that the copolymer thus produced also had the absorption relevant

to the carbon-carbon double bond at 1648cm^{-1} .

EXAMPLE J1

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5 A mixture composed of 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber containing the hydrolyzable silyl group (A-1), prepared in PRODUCTION EXAMPLE J1, 10 parts by weight of methanol (B-1) and 4 parts by weight of methyl orthoformate (B-2) was stirred 3 times at 10,000rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto Homogenizer), to prepare the composition. It was tested for storage stability at room temperature, after it was diluted to 35% by toluene and incorporated with 2,000ppm of moisture. The results are given in Table J2 for the test piece stored for 3 weeks.

REFERENCE EXAMPLE J1

15 Methyl dichlorosilane, 1.6mL and chloroplatinic acid, 0.00001g were added to 20g of the toluene solution of the acrylic-based polymer, prepared in PRODUCTION EXAMPLE J2, and they were allowed to react with each other at 90°C for 3 hours under sealed conditions. The effluent was incorporated with 20 5mL of methanol and 5mL of methyl orthoformate, and the mixed solution was continuously stirred until the solution became neutral.

The hydrosilylated product had an infrared absorption spectral pattern in which the infrared absorption at 1648cm^{-1} had

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completely disappeared.

The gas chromatography analysis results of the polymer solution are given in Table J1.

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Table J1

Gas chromatographic analysis results of the polymer solution	
Methyltrimethoxy silane	3.8%
Methyl orthoformate	5.0%
Methanol	10.5%

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The polymer solution thus prepared was tested for storage stability at room temperature, after it was diluted to 35% by toluene and incorporated with 2,000ppm of moisture. The results are given in Table J2 for the test piece stored for 3 weeks.

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Table J2

	Viscosity * (23°C ; cPs)		Viscosity changes (21 days/Initial)
	Initial	After 21 days	
EXAMPLE J1	300	520	1.7
REFERENCE EXAMPLE J1	10	21	2.1

*: Viscosity was determined by a B type viscometer at 23°C

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As shown in Table J2, the compositions prepared in EXAMPLE J1 and REFERENCE EXAMPLE J1 are excellent in storage stability.

REFERENCE EXAMPLE J2

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5 The hydrosilylation was effected in the same manner as in REFERENCE EXAMPLE J1, except that 1.6mL of methyl dichlorosilane was replaced by 1.8mL of methyl diethoxysilane. The hydrosilylated product also had an infrared absorption spectral pattern in which the infrared absorption at 1648cm^{-1} had completely disappeared, from which it was judged that
10 silyl-containing acrylic-based polymer was produced.

REFERENCE EXAMPLES J3 and J4

15 The hydrosilylation was effected in exactly the same manner as in REFERENCE EXAMPLE J1 for each of the above examples, except that the copolymer rubber (A-1) was replaced by the polymer prepared in respective PRODUCTION EXAMPLE J3 and J4, to prepare the resin curable at normal temperature.

REFERENCE EXAMPLES J5 to J8

20 Two parts by weight of dibutyl tin maleate was added to 100 parts by weight of the resin prepared in each of REFERENCE EXAMPLES J1 to J4, and each composition was spread over a soft steel plate, to evaluate its capacity of forming a coating film and other characteristics. The results are given in Table J3.

Table J3

Resin	Tack-free Time [minutes]	Time for which the test piece was left * [hrs]	Surface gloss *
EXAMPLE J1	20	48	Excellent
REFERENCE EXAMPLE J1	30	48	Excellent
* *	35	48	Excellent
REFERENCE EXAMPLE J2	40	48	Excellent
REFERENCE EXAMPLE J3	30	48	Excellent
REFERENCE EXAMPLE J4	45	72	Excellent

* : Left at 25°C and RH 70%

** : Resin prepared in REFERENCE EXAMPLE J1
which was further incorporated with 30% by
weight of ethyl silicate

Moreover, each composition prepared in EXAMPLE J1 and
REFERENCE EXAMPLES J5 to J8 was tested for curing speed, and
the cured product for resistance to weather, in accordance with
the methods described earlier. The results are given in Table
5 J4.

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Table J4

	Curing speed	Resistance to weather
EXAMPLE J1	○	No cracks observed
REFERENCE EXAMPLE J5	×	Cracks observed slightly
REFERENCE EXAMPLE J6	×	Cracks observed slightly
REFERENCE EXAMPLE J7	×	Cracks observed slightly
REFERENCE EXAMPLE J8	×	Cracks observed slightly

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<EXAMPLES K Series>

The composition, iodine value, intrinsic viscosity $[\eta]$ and molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and REFERENCE EXAMPLES were determined
5 by the methods described earlier.

The curing speed tests and accelerated weather resistance tests were conducted by the following methods for EXAMPLES and REFERENCE EXAMPLES.

(1) Curing speed test

10 The curable composition (stock material) was cured under the conditions of 23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

Next, the cured product was released from the mold, and thickness of the cured portion was measured by a dial gauge of
15 weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was 5mm or more, and × when it was less than 5mm.

(2) Accelerated weather resistance test

The weather resistance test was conducted in accordance with
20 JIS B-7753 using a Sunshine Carbon Arc weatherometer.

<Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

PRODUCTION EXAMPLE K1

[Production of silyl-containing
5 ethylene/propylene/5-vinyl-2-norbornene random copolymer
rubber (A-1)]

10 The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
propylene and 5-vinyl-2-norbornene were continuously supplied
at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$
and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
15 per hour, respectively, also continuously.

The copolymerization effected under the above conditions
produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber (A₀-1) in a form of uniform solution.

20 A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
the polymerization. The polymer was separated from the solvent
by steam-stripping the solution, and dried at 55°C for 48 hours
under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random

copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g measured in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and Mw/Mn of 15.

5 Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the
10 solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group ($-\text{SiCH}_3(\text{OCH}_3)_2$).

15 PRODUCTION EXAMPLE K2

[Production of the saturated hydrocarbon-based polymer]

p-DCC, 7.5mmols, represented by the following formula, compound A, was charged in a 1L pressure-resistant glass autoclave, to which agitator blades, a 3-way cock and vacuum
20 line were attached. Then, the autoclave was purged by nitrogen.

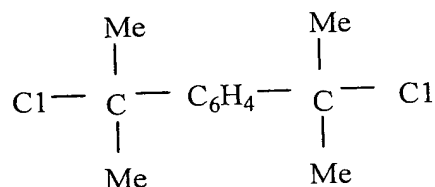
Then, the autoclave was charged with 330mL of toluene and 141mL of hexane, dried by a molecular sieve, as the solvents by a syringe, and then with 3.0mmols of α -picoline, while supplying nitrogen through one side of the 3-way cock.

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Next, a pressure-resistant glass-made liquefied gas collecting tube equipped with a needle valve and containing 113g of isobutylene, passed through a column packed with barium oxide for dehydration, was connected to the 3-way cock. Then the
5 autoclave as the polymerization reactor was immersed in a dry ice/acetone bath kept at -70°C , to cool the solution for 1 hour while stirring inside. The polymerization reactor was evacuated to a vacuum via the vacuum line, after it was cooled, and charged with isobutylene from the liquefied gas collecting
10 tube by opening the needle valve. Then, the reactor inside was returned back to the normal pressure by introducing nitrogen with handling the 3-way cock.

15 TiCl_4 , 7.18g (3.8mmols) was charged in the polymerization reactor by a syringe via the 3-way cock, after confirming that the reactor inside was kept at -70°C , to initiate the polymerization. After a lapse of 2 hours, 2.57g (22.5mmols) of allyl trimethylsilane was added to the reactor. The reaction process was continued for another 1 hour, and water was added to the reaction mixture to deactivate the catalyst. Then, the
20 organic layer was washed with pure water 3 times, and, after water was removed, distilled to remove the solvent under a vacuum. This produced the isobutylene polymer with the allyl group at the terminal.

Compound A is represented by the following structural formula:

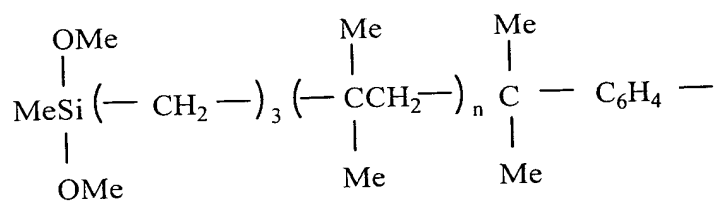


5 Then, 100g of the isobutylene polymer with the allyl group at the terminal was dissolved in 50mL of n-heptane, and the solution was heated to around 70°C, to which 1.2 [eq./allyl group] of methyl dimethoxysilane and 1×10^{-4} [eq./allyl group] of platinum (vinyl siloxane) complex were added for the

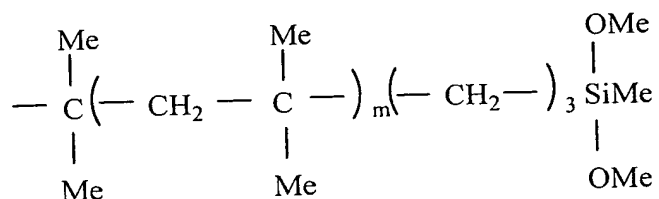
10 hydrosilylation. The reaction process was followed by FT-IR, and stopped in around 4 hours, after confirming that the olefin-derived absorption at 1640cm^{-1} disappeared.

The reaction solution was enriched under a vacuum, to produce the isobutylene polymer with the reactive silicon at both

15 terminals, represented by the following formula:



20



The polymer yield was estimated from the quantity produced. It was also analyzed for number-average molecular weight (Mn) and Mw/Mn by GPC, and the terminal structure by comparing the intensities of the ^1H -NMR-analyzed resonance signals of proton relevant to each structure (proton derived from the initiator: 6.5 to 7.5ppm, methyl proton bonded to the silicon atom, derived from the polymer terminal: 0.0 to 0.1ppm, and methoxy proton: 3.5 to 3.4) with each other. The polymer had an Mn of 11,416, Mn/Mw of 1.47 and Fn (silyl) of 1.95 (number-average molecular weight is a relative value to that of the standard polystyrene, and Fn (silyl) is number of the terminal functional silyl groups in one molecule of the isobutylene polymer).

EXAMPLE K1

A mixture containing the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE K1, was prepared. It was composed of 100 parts of the copolymer rubber (A-1), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320BTM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM) as Aging Inhibitor 1, 1 part of another aging inhibitor

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5 (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part
of still another aging inhibitor (Sankyo, Sanol LS-765TM) as
Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku
Kogyo, Sandant NBCTM), 3 parts of light-curable resin (TOAGOSEI,
5 Aronix M-400TM), 5 parts of a thixotropy imparting agent
(Kusumoto Kasei, Disparlon #305TM), and 4 parts of γ -isocyanate
propyltriethoxysilane as the silane coupling agent (Nippon
Unicar, Y-9030TM), all parts by weight. The mixture was well
kneaded by a 3-paint roll unit, to produce the major ingredient
10 for REFERENCE EXAMPLE K1.

The curing agent was prepared for EXAMPLE K1 by the following
procedure: a mixture comprising 4 parts of dibutyl tin
bisacetylacetonate (NITTO KASEI, U-220TM) as the silanol
condensing catalyst, 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts
15 of a paraffin-based process oil (Idemitsu Kosan, Diana Process
Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium,
Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical,
CB#30TM), all parts by weight, was manually kneaded in a disposal
cup and stirred 3 times at 10,000 rpm each for 10 minutes by
20 a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto
Homogenizer).

The above composition put in a sealed glass bottle was stored
for a month in a perfect oven kept at 50°C, to measure viscosity
of the major ingredient.

The viscosity was measured by a B type viscometer (Tokyo Keiki, Model BS) at 23°C using a No.7 rotor.

It had a viscosity of 8,525 poise at 10 rpm immediately after it was prepared, and 9,020 poise also at 10 rpm after it was stored.

REFERENCE EXAMPLES K1 and K2

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A mixture containing the polymer prepared in PRODUCTION EXAMPLE K2 was prepared for each of REFERENCE EXAMPLES K1 and K2. It was composed of 100 parts of the polymer, 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320BTM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM) as Aging Inhibitor 1, 1 part of another aging inhibitor (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part of still another aging inhibitor (Sankyo, Sanol LS-765TM) as Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku Kogyo, Sandant NBCTM), 3 parts of light-curable resin (TOAGOSEI, Aronix M-400TM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), and 4 parts of γ -isocyanate propyltriethoxysilane as the silane coupling agent (Nippon Unicar, Y-9030TM), all parts by weight. The mixture was well kneaded by a 3-roll unit, to produce the major ingredient

for REFERENCE EXAMPLE K1.

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10 The curing agent was prepared for REFERENCE EXAMPLE K1 by the following procedure: a mixture comprising 4 parts of dibutyl tin bisacetylacetonate (NITTO KASEI, U-220TM) as the silanol
5 condensing catalyst, 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal
10 cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto Homogenizer).

15 The major ingredient and curing agent were prepared for REFERENCE EXAMPLE K2 in the same manner as in REFERENCE EXAMPLE K1, except that 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) was incorporated in the former and omitted from the latter, to prepare the composition to be tested in the same manner.

20 Each of the above compositions put in a sealed glass bottle was stored for a month in a perfect oven kept at 50°C, to measure viscosity of the major ingredient.

The viscosity was measured by a B type viscometer (Tokyo Keiki, Model BS) at 23°C using a No. 7 rotor.

These compositions had a viscosity of 7,632 poise (REFERENCE EXAMPLE K1) and 8,928 poise (REFERENCE EXAMPLE K2) at 10 rpm

immediately after they were prepared, and 9,072 poise and higher than 12,000 poise (beyond the measurable range), respectively, also at 10 rpm after they were stored. The results indicate that the major ingredient has a higher viscosity when

5 incorporated with a hydrate of metallic salt than when not, and that its viscosity increases when it is stored.

EXAMPLE K2

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A mixture containing the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE K1, was prepared. It was composed of 100 parts of the copolymer rubber (A-1), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320TM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM) as Aging Inhibitor 1, 1 part of another aging inhibitor (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part of still another aging inhibitor (Sankyo, Sanol LS-765TM) as
15 Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku Kogyo, Sandant NBCTM), 3 parts of light-curable resin (TOAGOSEI, Aronix M-400TM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), 4 parts of γ -isocyanatepropyltriethoxysilane as the silane coupling agent
20

(Nippon Unicar, Y-9030TM) and 2 parts of γ -glycidoxypropyltrimethoxysilane (Nippon Unicar, A-187TM), all parts by weight. The mixture was well kneaded by a 3-paint roll unit, to produce the major ingredient for REFERENCE EXAMPLE K2.

5 The curing agent was prepared for EXAMPLE K2 by the following procedure: a mixture comprising 4 parts of dibutyl tin bisacetylacetonate (NITTO KASEI, U-220TM) as the silanol condensing catalyst, 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto
10 Homogenizer).
15

The above composition put in a sealed glass bottle was stored for a month in a perfect oven kept at 50°C, to measure changes with time in adhesion to various base materials and mechanical characteristics immediately after it was prepared and after it
20 was stored. The results are given in Tables K1 and K2.

The test piece for the tensile test was prepared in accordance with JIS A-5758/1992 specifying the method for preparing the test piece for the tensile test; the composition comprising the major ingredient and curing agent in a given weight ratio was

well kneaded and put in the H-shape of glass or aluminum substrate, and cured in an oven.

The curing conditions were 23°C×7 days + 50°C×7 days for each composition.

5 Three types of materials were used to prepare substrates for the H-type tensile test; float glass (Koen-sha, designated by Japan Sealant Industry Association, 3 by 5 by 0.5cm in size) in accordance with JIS A-5758/1992, pure aluminum (Taiyu Kizai, A1100P, 5 by 5 by 0.2cm in size) in accordance with JIS H-4000, and heat ray reflective glass (KLS™, 5 by 5 by 0.6cm). Each
10 of these H-shapes was washed with methylethylketone (Wako-Junyaku Kogyo, special grade) and wiped with clean cotton cloth, before it was filled with the composition.

The H-shape test piece thus prepared was tested by the method
15 of testing tensile adhesion in accordance with JIS A-5758/1992, wherein it was cured at a tensile speed of 50mm/minute in a constant-temperature chamber kept at 23°C and 65±5%RH. The cohesion fracture (CF)/thin film fracture (TCF)/adhesion fracture (AF) ratio shown in the tables for the K Series were
20 determined by visual observation of the cross-sections of the tensile-tested pieces.

REFERENCE EXAMPLES K3 and K4

A mixture containing the polymer prepared in PRODUCTION EXAMPLE K2 was prepared for each of REFERENCE EXAMPLES K3 and

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5 K4. It was composed of 100 parts of the polymer, 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320BTM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM) as Aging Inhibitor 1, 1 part of another aging inhibitor (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part of still another aging inhibitor (Sankyo, Sanol LS-765TM) as Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku Kogyo, Sandant NBCTM), 3 parts of light-curable resin (TOAGOSEI, Aronix M-400TM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), 4 parts of γ -isocyanate propyltriethoxysilane as the silane coupling agent (Nippon 10 Unicar, Y-9030TM), and 2 parts of γ -glycidoxypentyltrimethoxysilane (Nippon Unicar, A-187TM), all parts by weight. The mixture was well kneaded by a 3-roll unit, to produce the major ingredient for REFERENCE EXAMPLE K3.

20 The curing agent was prepared for REFERENCE EXAMPLE K3 by the following procedure: a mixture comprising 4 parts of dibutyl tin bisacetylacetonate (NITTO KASEI, U-220TM) as the silanol condensing catalyst, 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium,

Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto
5 Homogenizer).

The major ingredient and curing agent were prepared for REFERENCE EXAMPLE K4 in the same manner as in REFERENCE EXAMPLE K3, except that 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) was incorporated in the major ingredient of REFERENCE EXAMPLE K3 and omitted from the curing agent of REFERENCE EXAMPLE K3, to
10 prepare the composition to be tested in the same manner.

Each of the above compositions put in a sealed glass bottle was stored for a month in a perfect oven kept at 50°C, to measure changes with time in adhesion to various base materials and mechanical characteristics immediately after it was prepared
15 and after it was stored. The results are given in Tables K1 and K2.

The test piece for the tensile test was prepared in accordance with JIS A-5758/1992 specifying the method for preparing the
20 test piece for the tensile test; the composition comprising the major ingredient and curing agent in a given ratio was well kneaded and put in the H-shape of glass or aluminum substrate, and cured in an oven.

The curing conditions were 23°C×7 days + 50°C×7 days for each

composition.

Three types of materials were used to prepare substrates for the H-type tensile test; float glass (Koen-sha, designated by Japan Sealant Industry Association, 3 by 5 by 0.5cm in size) in accordance with JIS A-5758/1992, pure aluminum (Taiyu Kizai, A1100P, 5 by 5 by 0.2cm in size) in accordance with JIS H-4000, and heat ray reflective glass (KLSTM, 5 by 5 by 0.6cm in size). Each of these H-shapes was washed with methylethylketone (Wako-Junyaku Kogyo, special grade) and wiped with clean cotton cloth, before it was filled with the composition.

The H-shape test piece thus prepared was tested by the method of testing tensile adhesion in accordance with JIS A-5758/1992, wherein it was cured at a tensile speed of 50mm/minute in a constant-temperature chamber kept at 23°C and 65±5%RH. The cohesion fracture (CF)/thin film fracture (TCF)/adhesion fracture (AF) ratio shown in the tables for the K Series were determined by visual observation of the cross-sections of the tensile-tested pieces.

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Table K1 (H type tensile test results with the just produced compositions)

	Substrate	M ₅₀ kgf/cm ²	T _B kgf/cm ²	E _B %	Fractured conditions (%)		
					CF	TCF	AF
EXAMPLE K2	FL	6.22	6.60	68	100	0	0
	pAl	5.10	6.80	72	100	0	0
	KLS	5.20	6.61	68	100	0	0
REFERENCE EXAMPLE K3	FL	6.14	7.80	79	100	0	0
	pAl	5.56	7.96	88	100	0	0
	KLS	5.97	7.79	82	99	1	0
REFERENCE EXAMPLE K4	FL	5.48	7.49	85	99	1	0
	pAl	4.94	8.13	107	94	5	1
	KLS	5.39	7.82	95	99	1	0

(Notes)

FL: Float glass, pAl: pure aluminum, KLS: Heat ray reflective glass, M₅₀: 50% Tensile stress,T_B: Tensile breaking strength, E_B: Tensile breaking elongation

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Table K2 (H type tensile test results with the stored compositions)

	Substrate	M ₅₀	T _B	E _B	Fractured conditions (%)		
		kgf/cm ²	kgf/cm ²	%	CF	TCF	AF
EXAMPLE K2	FL	5.20	7.10	79	100	0	0
	pAl	5.12	7.32	85	98	2	0
	KLS	5.24	7.06	77	98	2	0
REFERENCE EXAMPLE K3	FL	5.94	8.88	91	100	0	0
	pAl	5.47	8.39	93	97	0	3
	KLS	6.50	9.01	81	98	2	0
REFERENCE EXAMPLE K4	FL	-	2.62	30	1	0	99
	pAl	-	1.85	23	0	0	100
	KLS	-	1.32	15	0	0	100

(Notes)

FL: Float glass, pAl: pure aluminum, KLS: Heat ray reflective glass, M₅₀: 50% Tensile stress,T_B: Tensile breaking strength, E_B: Tensile breaking elongation

EXAMPLE K3

The curing agent was prepared in the same manner as in EXAMPLE K2, except that U-220 as the silanol condensing catalyst was replaced by 4 parts by weight of dibutyl tin dimethoxide (Aldrich Chemical), and tested also by use of the major ingredient of EXAMPLE K2 in the same manner as in EXAMPLE K2. The results are given in Tables K3 and K4.

REFERENCE EXAMPLE K5

The curing agent was prepared in the same manner as in REFERENCE EXAMPLE K3, except that U-220 as the silanol condensing catalyst was replaced by 4 parts by weight of dibutyl tin dimethoxide (Aldrich Chemical), and tested also by use of the major ingredient of REFERENCE EXAMPLE K3 in the same manner as in REFERENCE EXAMPLE K3. The results are given in Tables K3 and K4.

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Table K4 (H type tensile test results with the stored compositions)

	Substrate	M ₅₀ kgf/cm ²	T _B kgf/cm ²	E _B %	Fractured conditions (%)			
					CF	TCF	AF	
EXAMPLE K3	FL	3.86	7.09	116	100	0	0	
	pAl	3.76	7.22	122	99	1	0	
	KLS	3.86	7.33	126	100	0	0	
REFERENCE EXAMPLE K5	FL	4.50	8.34	133	98	0	2	
	pAl	4.32	8.46	139	100	0	0	
	KLS	4.69	8.41	125	100	0	0	

(Notes)

FL: Float glass, pAl=pure aluminium, KLS: Heat ray reflective glass, M₅₀: 50% Tensile stress,T_B: Tensile breaking strength, E_B: Tensile breaking elongation

EXAMPLES K4 to K7

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A mixture containing the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE K1, was prepared for each of EXAMPLES K4 to K7. It was composed of 100 parts of the copolymer rubber (A-1), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320BTM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM) as Aging Inhibitor 1, 1 part of another aging inhibitor (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part of still another aging inhibitor (Sankyo, Sanol LS-765TM) as Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku Kogyo, Sandant NBCTM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), 4 parts of the silane coupling agent 1 (Nippon Unicar, Y-9030TM) and 2 parts of the silane coupling agent 2 (Nippon Unicar, A-187TM), all parts by weight. The mixture was well kneaded by a 3-paint roll unit, to produce the major ingredient for REFERENCE EXAMPLES K6 to K9.

The curing agent was prepared for EXAMPLE K4 by the following procedure: a mixture comprising 4 parts of the silanol condensing catalyst (NITTO KASEI, U-220TM), 4 parts of salt cake

($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd. Excel Auto Homogenizer).

The composition was prepared for each of EXAMPLES K4 to K7 in the same manner as in EXAMPLE K2, except that salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) for the curing agent was replaced by another hydrate of metallic salt, 6 parts of hypo ($\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$) for EXAMPLE K5, 6 parts of magnesium sulfate ($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$) for EXAMPLE K6 and 4 parts of sodium phosphate ($\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$) for EXAMPLE K7 to prepare the curing agent, and tested in the same manner as in EXAMPLE K2. The results are given in Tables K5 and K6.

REFERENCE EXAMPLES K6 to K9

A mixture containing the polymer prepared in PRODUCTION EXAMPLE K2 was prepared for each of REFERENCE EXAMPLES K6 to K9. It was composed of 100 parts of the polymer, 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 180 parts of limestone powder (Shiraishi Calcium, PO320BTM), 50 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Maruo Calcium, LMRTM), 1 part of an aging inhibitor (Ciba-Geigy Japan, Irganox 1010TM)

as Aging Inhibitor 1, 1 part of another aging inhibitor (Sumitomo Chemical, Sumisorb 400TM) as Aging Inhibitor 2, 1 part of still another aging inhibitor (Sankyo, Sanol LS-765TM) as Aging Inhibitor 3, 3 parts of a light stabilizer (Sanshin Kagaku Kogyo, Sandant NBCTM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), 4 parts of silane coupling agent 1 (Nippon Unicar, Y-9030TM) and 2 parts of silane coupling agent 2 (Nippon Unicar, A-187TM), all parts by weight. The mixture was well kneaded by a 3-paint roll unit, to produce the major ingredient.

The curing agent was prepared for REFERENCE EXAMPLE K6 by the following procedure: a mixture comprising 4 parts of the silanol condensing catalyst (NITTO KASEI, U-220TM), 4 parts of salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Seisakusho Co., Ltd. Excel Auto Homogenizer).

The composition was prepared for each of REFERENCE EXAMPLES K7 to K9 in the same manner as in REFERENCE EXAMPLE K3, except that salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$) for the curing agent was replaced by another hydrate of metallic salt; 6 parts of hypo ($\text{Na}_2\text{S}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$)

for REFERENCE EXAMPLE K7, 6 parts of magnesium sulfate
($\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$) for REFERENCE EXAMPLE K8 and 4 parts of sodium
phosphate ($\text{Na}_3\text{PO}_4 \cdot 12\text{H}_2\text{O}$) for REFERENCE EXAMPLE K9 to prepare the
curing agent, and tested in the same manner as in REFERENCE EXAMPLE
5 K3. The results are given in Tables K5 and K6.

The composition comprising the major ingredient and curing
agent, prepared in each of EXAMPLES K1 to K7 and REFERENCE EXAMPLES
K1 to K9 was tested for curing speed and resistance to weather
by the methods described earlier.

10 The results are given in Table K7.

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Table K6 (H type tensile test results with the stored compositions)

	Substrate	M ₅₀ kgf/cm ²	T _B kgf/cm ²	E _B %	Fractured conditions (%)		
					CF	TCF	AF
EXAMPLE K4	FL	4.33	6.25	84	100	0	0
	KLS	4.34	6.34	91	100	0	0
EXAMPLE K5	FL	4.87	6.70	80	100	0	0
	KLS	4.88	6.80	77	100	0	0
EXAMPLE K6	FL	4.58	7.00	92	100	0	0
	KLS	4.54	7.42	94	100	0	0
EXAMPLE K7	FL	4.22	6.80	95	100	0	0
	KLS	4.32	6.90	102	100	0	0
REFERENCE EXAMPLE K6	FL	4.92	7.40	90	100	0	0
	KLS	5.20	7.29	83	99	1	0
REFERENCE EXAMPLE K7	FL	5.51	7.71	80	100	0	0
	KLS	5.63	7.45	74	100	0	0
REFERENCE EXAMPLE K8	FL	5.22	8.45	99	100	0	0
	KLS	5.31	7.94	87	100	0	0
REFERENCE EXAMPLE K9	FL	4.78	7.69	102	100	0	0
	KLS	5.12	7.76	96	100	0	0

(Notes)

FL: Float glass, KLS: Heat ray reflective glass, M₅₀: 50% Tensile stress,T_B: Tensile breaking strength, E_B: Tensile breaking elongation

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Table K7

	Just produced compositions		Stored compositions	
	Curing speed	Resistance to weather	Curing speed	Resistance to weather
EXAMPLE K1	○	No cracks or molten portion observed	○	No cracks or molten portion observed
REFERENCE EXAMPLE K1	×	No cracks or molten portion observed	×	No cracks or molten portion observed
REFERENCE EXAMPLE K2	×	No cracks or molten portion observed	×	No cracks or molten portion observed
EXAMPLE K2	○	No cracks or molten portion observed	○	No cracks or molten portion observed
REFERENCE EXAMPLE K3	×	No cracks or molten portion observed	×	No cracks or molten portion observed
REFERENCE EXAMPLE K4	×	No cracks or molten portion observed	×	No cracks or molten portion observed
EXAMPLE K3	○	No cracks or molten portion observed	○	No cracks or molten portion observed
REFERENCE EXAMPLE K5	×	No cracks or molten portion observed	×	No cracks or molten portion observed
EXAMPLE K4	○	No cracks or molten portion observed	○	No cracks or molten portion observed
EXAMPLE K5	○	No cracks or molten portion observed	○	No cracks or molten portion observed
EXAMPLE K6	○	No cracks or molten portion observed	○	No cracks or molten portion observed
EXAMPLE K7	○	No cracks or molten portion observed	○	No cracks or molten portion observed
REFERENCE EXAMPLE K6	×	No cracks or molten portion observed	×	No cracks or molten portion observed
REFERENCE EXAMPLE K7	×	No cracks or molten portion observed	×	No cracks or molten portion observed
REFERENCE EXAMPLE K8	×	No cracks or molten portion observed	×	No cracks or molten portion observed
REFERENCE EXAMPLE K9	×	No cracks or molten portion observed	×	No cracks or molten portion observed

<EXAMPLES L Series>

PRODUCTION EXAMPLE 1

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
5 rubber]

The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
10 propylene and 5-vinyl-2-norbornene were continuously supplied
at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$
and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
per hour, respectively also continuously.

15 The copolymerization effected under the above conditions
produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
20 the polymerization. The polymer was separated from the solvent
by steam-stripping the solution, and dried at 55°C for 48 hours
under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber thus produced contained ethylene at 68% by mol,

and had an iodine value of 10, an intrinsic viscosity $[\eta]$ of 0.2dl/g, and M_w/M_n of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the copolymer rubber containing dimethoxymethylsilyl group.

10 EXAMPLES L1 to L4, and COMPARATIVE EXAMPLE L1

A uniform rubber composition was prepared in each of EXAMPLES L1 to L4, and COMPARATIVE EXAMPLE L1 using the components given in Table L1 in a ratio also given in Table L1, to measure its viscosity. The results are shown in Table L1.

15 Each composition prepared was flow-cast to a 3mm thick sheet, and cured at room temperature for 4 days and 50°C for another 4 days.

The cured sheet was evaluated for its tackiness, resistance to heat, curing speed and resistance to weather by the following methods.

20 Curing speed (film expandability) and resistance to weather were also measured using different aliquots of each example. The results are given in Table L2.

1) Tackiness

Tackiness was evaluated according to the following three-grade system by feeling of touching the sheet with a finger:

- ◎: No tackiness is felt to the touch
 - : Tackiness is felt to the touch slightly
 - 5 △: Tackiness is felt to the touch
- 2) Resistance to heat

Resistance to heat was evaluated by the time required for the sheet surface to start melting at 130°C.

3) Resistance to weather

- 10 Resistance to weather was evaluated by the time required for the sheet surface to start melting in the accelerated weather resistance test using a Sunshine weatherometer.

4) Curing speed test

- 15 The composition comprising the major ingredient and the catalyst was measured for its film expandability at room temperature, i.e., curing speed.

- 20 The composition and mold (20 by 80 by 5mm in size) were kept and adjusted under the conditions of 23°C and 50%RH overnight, and then the mold was filled with the composition. Next, the cured composition was released from the mold after it was kept therein for 24 hours, and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed.

(Evaluation standards)

× : when thickness of the cured portion is less than 1mm

○ : when thickness of the cured portion is 1mm or more

5) Weather resistance test

The accelerated weather resistance test was conducted in
5 accordance with JIS B-7753:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120
minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

10 Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

The surface state of tested test piece was visually observed,
to evaluate its resistance to weather according to the following
three grades:

15 ○ : No cracks or molten portion observed

△ : Cracks or molten portion observed slightly

× : Cracks or molten portion observed

REFERENCE EXAMPLES L1 to L6

Three types of polymers containing a reactive silicon group
20 were prepared by the methods disclosed by Japanese Patent
Laid-Open Publication No.252670/1989 in PRODUCTION EXAMPLES 1
to 3 (column 16 to 18).

A uniform rubber composition was prepared using each of these
polymers and other components shown in Tale L1. Table L1

describes the ratio of these compositions.

These rubber compositions and those cured products prepared in EXAMPLES were evaluated for the properties in the same manner as in EXAMPLES. The results are given in Table L1.

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Table L1

Composition (Parts)	EXAMPLE L1	EXAMPLE L2	REFERENCE EXAMPLE L1	REFERENCE EXAMPLE L2	EXAMPLE L3	EXAMPLE L4	REFERENCE EXAMPLE L3	REFERENCE EXAMPLE L4	COMPARATIVE EXAMPLE L1	REFERENCE EXAMPLE L5	REFERENCE EXAMPLE L6
Component (A2) PRODUCTION EXAMPLE L1 PRODUCTION EXAMPLE L2 (Note 6) PRODUCTION EXAMPLE L3 (Note 7)	80 - -	80 - -	- 80 -	- 80 -	100 - -	100 - -	- - 100	- - 100	100 - -	- 100 -	- - 100
Component (K1) PS 340.5 (Note 1) PS 084 (Note 2) PS 080 (Note 3)	20 - -	- 20 -	20 - -	- 20 -	- - 24 (Note 4)	- - 35 (Note 5)	- - 80 (Note 4)	- - 90 (Note 5)	- - -	- - -	- - -
Tin octylate	3	3	3	3	3	3	3	3	3	3	3
Lauryl amine	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75	0.75
Curability (day, 23°C)	○	○	×	×	○	○	×	×	○	×	×
Tackiness	◎	◎	◎	◎	◎	◎	◎	◎	○	△	○
Resistance to heat (day)	110	105	45	48	120	115	40	41	115	50	43
Resistance to weather (day)	○	○	△	△	○	○	△	△	○	△	△

Notes for Table L1:

(Note 1) PS 340.5: Polydimethylsiloxane with a silanol group at the terminal (Chisso Corp.)

5 (Note 2) PS 084: Polydimethyldiphenylsiloxane with a diphenyl silanol group at the terminal (Chisso Corp.)

(Note 3) PS 080: Polydiphenylsiloxane with a silanol group at the terminal (Chisso Corp.)

(Note 4) Ratio of silanol group in polysiloxane to methoxysilyl group in the polymer: 1 by equivalent

10 (Note 5) Ratio of silanol group in polysiloxane to methoxysilyl group in the polymer: 1.2 by equivalent

(Note 6) Polymer with a reactive silicon group, synthesized in accordance with the methods disclosed by Japanese Patent Laid-Open Publication No.252670/1989 in PRODUCTION EXAMPLE 1 (column 16 to 18).

(Note 7) Polymer with a reactive silicon group, synthesized in accordance with the methods disclosed by Japanese Patent Laid-Open Publication No.252670/1989 in PRODUCTION EXAMPLE 3 (column 16 to 18).

20 <EXAMPLES M Series>

PRODUCTION EXAMPLE M1

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber thus produced contained ethylene at 68% by mol, and had an iodine value of 10, an intrinsic viscosity $[\eta]$ of 0.2dl/g, and M_w/M_n of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer

rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the random copolymer rubber containing dimethoxymethylsilyl group.

EXAMPLES M1 to M5

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A rubber composition was prepared in each of EXAMPLES M1 to M5 using the components given in Table M1 in a ratio also given in Table M1. It comprised the dimethoxymethylsilyl group containing copolymer rubber prepared in PRODUCTION EXAMPLE M1 as the component (A2); and an organic rubber of polybutadiene rubber, styrene/butadiene copolymer rubber, acrylic rubber (JSR Corp., AR101), polypropylene glycol containing a hydrolyzable silyl group (KANEKA CORP., MS Polymer, MS203™) or nitrile rubber (JSR Corp., N230S) as the component (K2). The mixture was kneaded by a Banbury mixer (Kobe Steel, Ltd. 1.9) at 120°C for 5 minutes, and further by an 8-inch open roll in the presence of a vulcanizing agent. The sample of the rubber composition was measured for vulcanizing speed T_{90} in accordance with JIS K-6300/1994.

Moreover, it was extruded into a sheet by a biaxial extruder, and continuously vulcanized under heating at 180°C for 1 hour, to produce the vulcanized rubber sheet.

The cured sheet was evaluated for tensile elongation,

resistance to heat, surface resistance to weather, curing speed and resistance to weather by the following methods. The results are given in Table M1.

(Evaluation methods)

5 1) Tensile test

The tensile test was conducted in accordance with JIS 6251 at 23°C using the JIS No.1 dumbbell-shaped test piece.

2) Weather resistance test

10 The accelerated weather resistance test was conducted in accordance with JIS B-7753, to determine resistance to weather:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

15 Tank inside temperature: 40±2°C

Total light irradiation time: 500 hours

The surface state of tested test piece was visually observed, to evaluate its resistance to weather according to the following three grades:

20 ○ : No cracks or molten portion observed

△ : Cracks or molten portion observed slightly

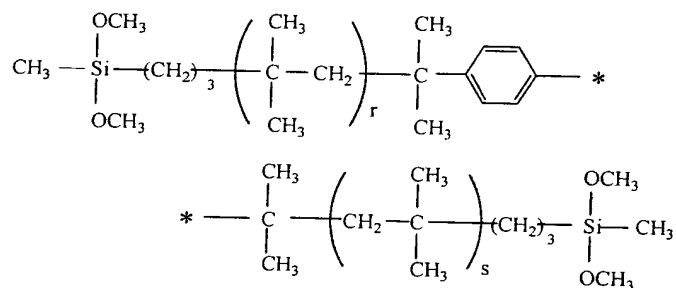
× : Cracks or molten portion observed

REFERENCE EXAMPLES M1 to M5

The vulcanized rubber sheet was prepared and tested in each

of REFERENCE EXAMPLES M1 to M5 as shown in Table M1 in the same manner as in corresponding EXAMPLE, except that the silyl-containing copolymer rubber as the component (A2) was replaced by the compound A (represented by the following general formula), synthesized by the method disclosed by Japanese Patent Laid-Open Publication No.105005/1988, and having a number-average molecular weight of 10,600, a molecular weight distribution (Mw/Mn) of 1.2 and terminal functional dimethoxymethylsilyl group number of 1.9. The results are given in Table M1.

Compound A



wherein, "r" and "s" are each an integer.

Table M1

	EXAMPLES					REFERENCE EXAMPLES				
	M1	M2	M3	M4	M5	M1	M2	M3	M4	M5
Composition (parts by weight)										
Component (A2)	50	50	50	50	50					
Silyl-containing copolymer rubber compound A										
Component (K2)						100	100	100	100	100
Polybutadiene rubber										
Styrene/butadiene copolymer rubber										
Acrylic rubber										
MS Polymer, MS203										
Nitrile rubber										
Other component										
Asahi#60G	40	40	40	40	40	40	40	40	40	40
Vulcanizer (M)										
Dicumyl peroxide	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.7	2.7
Curing speed of rubber composition: T90 (170°C) minutes	5.6	5.8	6.2	6.9	6.5	6.8	6.5	8.2	10.5	8.6
Properties of the cured product	400	420	480	500	490	250	280	290	300	310
Tensile elongation (%)	○	○	○	○	○	△	△	△	△	△
Resistance to weather										

Notes)

MS Polymer MS203™: Polypropylene glycol containing a hydrolysable silyl group, KANEKA Corp.

Asahi #60G: FEF grade carbon black, Asahi Carbon

As shown in Table M1, the rubber composition prepared in each of EXAMPLES is superior to that prepared in corresponding REFERENCE EXAMPLE in all of curing speed, and surface resistance to weather, resistance to weather and resistance to heat in its cured product.

<EXAMPLES N Series>

PRODUCTION EXAMPLE 1

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate

the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random
5 copolymer rubber thus produced contained ethylene at 68% by mol, and had an iodine value of 10, an intrinsic viscosity $[\eta]$ of 0.2dl/g, and M_w/M_n of 15.

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10 Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the random copolymer rubber containing dimethoxymethylsilyl
15 group.

REFERENCE PRODUCTION EXAMPLES 1 to 6

The polymer was prepared in each of REFERENCE PRODUCTION EXAMPLES 1 to 6 in accordance with the respective method of PRODUCTION EXAMPLES 1 to 6 disclosed by Japanese Patent Laid-Open
20 Publication No.292616/1992 [0036] to [0037].

EXAMPLES N1 to N4, and REFERENCE EXAMPLES N1 to N6

The curable resin composition was prepared in each of EXAMPLES N1 to N4 using the components given in Table N1 in a weight ratio also given in Table N1. It comprised the copolymer rubber

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containing dimethoxysilyl group, prepared in PRODUCTION EXAMPLE 1, an epichlorohydrin-bisphenol A type epoxy resin (Yuka Shell Epoxy, Epikote #828TM, epoxy equivalents: around 190), γ -(2-aminoethyl) aminopropyl trimethoxysilane (Nippon Unicar, A1122TM) as the silane coupling agent, 50/50 mixture of dibutyl tin oxide and dioctyl phthalate (Sankyo Organic Chemicals, Ltd., #918TM) as the silanol condensing catalyst, and 2,4,6-tris(dimethylaminomethyl)phenol (Kayaku Nooly Co., Ltd., DMP30TM) as the epoxy resin curing catalyst. The composition was also prepared in the same manner as in the above, except that the polymers prepared in COMPARATIVE PRODUCTION EXAMPLES 1 to 6 were used for respective COMPARATIVE EXAMPLES N1 to N6.

The curable resin composition prepared in each of EXAMPLES N1 to N4, and COMPARATIVE EXAMPLES N1 to N6 was evaluated by the following methods.

1) Tensile test using the dumbbell-shaped test piece

The curable resin composition was molded in a Teflon frame into a sheet by curing at 23°C for 3 days and 50°C for another 4 days. The cured sheet was stamped out into the No.3 dumbbell-shaped test piece in accordance with JIS K-6301. It was stretched at 200mm/minute, to determine its moduli at 50 and 100% tension (M_{50} and M_{100}), breaking strength (T_B) and breaking elongation (E_B).

2) Measurement of tensile shear strength

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The test was conducted in accordance with JIS K-6850. An aluminum plate (A-1050P aluminum plate, 100 by 25 by 2mm in size, specified by JIS H-4000) was wiped lightly with acetone, on which the curable resin composition was spread by a spatula to an area of around 25 by 12.5mm and thickness of 0.05mm. The coated surfaces of the two plates were attached face-to-face and manually pressed against each other. The coated test pieces were fixed, and the resin was cured at 23°C for 3 days and 50°C for another 4 days. Then, they were stretched at 5mm/minute away from each other until their cured resin was fractured. The maximum load value measured at which the cured resin was fractured was divided by the coated area to find the tensile shear strength.

Curing speed and resistance to weather were measured by the following methods:

15 3) Curing speed

The composition comprising the major ingredient and the catalyst was measured for its film expandability at room temperature, i.e., curing speed.

20 The curable composition was cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm.

(Evaluation standards)

× : when thickness of the cured portion is less than 1mm

○ : when thickness of the cured portion is 1mm or more

4) Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753, to determine resistance to weather:

5 Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^{\circ}\text{C}$

Tank inside temperature: $40 \pm 2^{\circ}\text{C}$

10 Total light irradiation time: 500 hours

The surface state of tested test piece was visually observed, to evaluate its resistance to weather according to the following three grades:

○ : No cracks or molten portion observed

15 △ : Cracks or molten portion observed slightly

× : Cracks or molten portion observed

The results are given in Table N1.

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Table N1

	EXAMPLE N				REFERENCE EXAMPLE N					
	1	2	3	4	1	2	3	4	5	6
Silyl-containing polymers PRODUCTION (P) EXAMPLE 1	100	100	100	100	-	-	-	-	-	-
COMPARATIVE P. EXAMPLE 1	-	-	-	-	100	-	-	-	-	-
COMPARATIVE P. EXAMPLE 2	-	-	-	-	-	100	-	-	-	-
COMPARATIVE P. EXAMPLE 3	-	-	-	-	-	-	100	-	-	-
COMPARATIVE P. EXAMPLE 4	-	-	-	-	-	-	-	100	-	-
COMPARATIVE P. EXAMPLE 5	-	-	-	-	-	-	-	-	100	-
COMPARATIVE P. EXAMPLE 6	-	-	-	-	-	-	-	-	-	100
Epoxy resin #828	50	50	50	50	50	50	50	50	50	50
Silane coupling agent	1	2	5	7.5	2	2	2	2	2	2
Silanol condensing catalyst	1	1	1	1	1	1	1	1	1	1
Epoxy curing catalyst	5	5	5	5	5	5	5	5	5	5
M ₅₀	14.9	30.1	23.7	21.6	32.7	36.9	27.4	24.9	37.9	10.7
M ₁₀₀	29.7	55.4	51.3	50.7	59.6	64.4	46.3	41.8	67.1	21.2
T _B (kgf/cm ²)	78.1	86.3	80.1	77.7	90.5	77.7	74.4	112	121	68.2
E _R (%)	180	131	111	107	158	125	180	314	189	250
Shear strength (kgf/cm ²)	91	138	133	131	147	180	100	143	92	100
Curing speed	○	○	○	○	○	○	○	○	○	○
Resistance to weather	○	○	○	○	×	×	×	×	×	×

<EXAMPLES O Series>

PRODUCTION EXAMPLE 1

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
5 rubber]

The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
10 propylene and 5-vinyl-2-norbornene were continuously supplied
at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$
and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
per hour, respectively, also continuously.

15 The copolymerization effected under the above conditions
produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
20 the polymerization. The polymer was separated from the solvent
by steam-stripping the solution, and dried at 55°C for 48 hours
under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber thus produced contained ethylene at 68% by mol,

and had an iodine value of 10, intrinsic viscosity $[\eta]$ of 0.2dl/g, and Mw/Mn of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber containing dimethoxymethylsilyl group.

REFERENCE PRODUCTION EXAMPLES 1 and 2

The polymers were prepared in REFERENCE PRODUCTION EXAMPLES 1 and 2 in accordance with the respective methods of PRODUCTION EXAMPLES 1 and 3, respectively, disclosed by Japanese Patent Laid-Open Publication No.280217/1987.

EXAMPLE 01, and REFERENCE EXAMPLES 01 and 02

A mixture containing the copolymer rubber as the component (A2), prepared in PRODUCTION EXAMPLE 1, was prepared for EXAMPLE 01. It was composed of 100 parts of the copolymer rubber, 50 parts of a bisphenol A type epoxy resin (Yuka Shell Epoxy, Epikote #828TM), 1 part of a bisphenol type antioxidant (Ouchishinko Chemical Industrial Co., Nocrac NS-6TM), 1 part of N- β -(aminoethyl)- γ -aminopropyltrimethoxysilane (silicon compound as the component (Q) for the present invention), 1 part

of diphenyl silanediol (silicon compound as the component (R) for the present invention), 2 parts of an organotin-based compound (Sankyo Organic Chemicals, Ltd., #918TM) as the silanol condensing catalyst, 5 parts of

5 2,4,6-tris(dimethylaminomethyl) phenol as the epoxy resin curing agent, and 0.4 part of water, all by weight. These components were well mixed with each other, and the mixture was poured into a polyethylene frame carefully to prevent the bubbles from entering the frame, where it was cured at 23°C for 1 day and 50°C for another 3 days into the 3mm thick sheet. The similar
10 procedures were repeated for preparation of the mixtures and sheets containing the polymers prepared in REFERENCE PRODUCTION EXAMPLES 1 and 2 (REFERENCE EXAMPLES O1 and O2).

The composition prepared in each of EXAMPLE O1, and REFERENCE
15 EXAMPLES O1 and O2 was tensile-tested using the dumbbell-shaped test piece, and also measured for curing speed and resistance to weather by the following methods. The results are given in Table O1.

1) Tensile test using the dumbbell-shaped test piece

20 The cured sheet was stamped out into the No.3 dumbbell-shaped test piece in accordance with JIS K-6301. It was stretched at 500mm/minute, to determine its breaking strength (T_B) and breaking elongation (E_B).

Curing speed and resistance to weather were determined by

the following methods.

1) Curing speed

The composition comprising the major ingredient and the catalyst was measured for its film expandability at room
5 temperature, i.e., curing speed.

The curable composition prepared in each of EXAMPLE and REFERENCE EXAMPLES was cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and thickness of the cured portion was measured by a dial gauge of weak spring force to
10 0.1mm.

(Evaluation standards)

× : when thickness of the cured portion is less than 1mm

○ : when thickness of the cured portion is 1mm or more

2) Weather resistance test

15 The accelerated weather resistance test was conducted in accordance with JIS B-7753, to determine resistance to weather:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

20 Black panel temperature: 63±2°C

Tank inside temperature: 40±2°C

Total light irradiation time: 500 hours

The surface state of tested test piece was visually observed, to evaluate its resistance to weather according to the following

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three grades:

○ : No cracks or molten portion observed

△ : Cracks or molten portion observed slightly

× : Cracks or molten portion observed

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Table O1

	Tensile properties		Curing speed	Resistance to weather
	T _B (kg/cm ²)	E _B (%)		
EXAMPLE O1	35	380	○	○
REFERENCE EXAMPLE O1	48	410	○	×
REFERENCE EXAMPLE O2	25	210	×	×

EXAMPLES O2 to O4, and REFERENCE EXAMPLES O3 to O5

The cured sheet was prepared in the same manner as in EXAMPLE O1 for each of EXAMPLES O2 to O4, except that diphenyl silanediol as the component (R) for the present invention was replaced by 0.5 part of bis (hydroxydimethylsilyl) benzene and 0.5 part of polydimethyl-diphenylsiloxane with terminal diphenyl silanol group (Petrarch Systems Inc., PS-084TM) or 0.5 part of silicone varnish having silanol group (Shin-Etsu Chemical Co., Ltd., KR-212TM), all by weight, to measure T_B and E_B . The cured sheet was prepared similarly using the composition prepared in REFERENCE EXAMPLE O1 for each of REFERENCE EXAMPLES O3 to O5. The results are given in Table O2.

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Table O2

	Types of component (R)	Tensile properties		Curing speed	Resistance to weather
		T _R (kg/cm ²)	E _R (%)		
EXAMPLE O2	Bis(hydroxydimethylsilyl) benzene	67	550	○	○
EXAMPLE O3	PS-084	65	800	○	○
EXAMPLE O4	KR-212	73	720	○	○
REFERENCE EXAMPLE O3	Bis(hydroxydimethylsilyl) benzene	55	510	○	×
REFERENCE EXAMPLE O4	PS-084	55	750	○	×
REFERENCE EXAMPLE O5	KR-212	63	660	○	×

EXAMPLES 05 to 08, and REFERENCE EXAMPLES 07 to 012

The adhesion test piece was prepared from the composition prepared in each of EXAMPLES 01 to 04 by the following procedure, to measure its adhesion strength in EXAMPLES 05 to 08. The results are shown in Table 03.

- Methods of preparing the test piece and testing for the tensile shear strength (in accordance with JIS K-6850)

An aluminum plate (A-1050P aluminum plate, 100 by 25 by 2mm in size, specified by JIS H-4000) was wiped lightly with acetone, on which the resin composition was spread by a spatula to an area of around 25 by 12.5mm and thickness of 0.05mm. The coated surfaces of the two plates were attached face-to-face and manually pressed against each other. The coated test pieces were fixed, and the resin composition was cured at 23°C for 1 day and 50°C for another 3 days. Then, they were stretched at 5mm/minute away from each other until their cured resin was fractured. The maximum load value measured at which the cured resin was fractured was divided by the sheared area to find the tensile shear strength.

- Methods of preparing the test piece and testing for the T type releasing strength

An aluminum plate (A-1050P aluminum plate, 200 by 25 by 0.1mm in size, specified by JIS H-4000) was wiped lightly with acetone, on which the resin composition was spread by a spatula to an

area of around 100 by 25mm and thickness of 0.3mm. The coated surfaces of the two plates were attached face-to-face and pressed against each other by a 5kg hand roller repeatedly 5 times in such a way to avoid back-and-forth motion. The resin composition was cured at 23°C for 1 day and 50°C for another 3 days. Then, the test piece thus prepared was set in a tensile tester in a T-shape, and stretched at 200mm/minute until the adhesive portion was fractured. The strength at which the adhesive portion was fractured was measured as the T-type releasing strength.

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Table O3

		Types of compositions used	Adhesion strength	
			Tensile shear Strength (kg/cm ²)	T type releasing strength (kg/25 mm)
EXAMPLES	O5	EXAMPLE O1	142	13.2
	O6	EXAMPLE O2	116	10.7
	O7	EXAMPLE O3	104	11.1
	O8	EXAMPLE O4	105	8.9

EXAMPLE 09

The curable resin composition was prepared in EXAMPLE 01 using the polymer prepared in PRODUCTION EXAMPLE 1. It comprised 100 parts of the polymer, 50 parts of Epikote #828TM, 1 part of Nocrac NS-6TM, 1 part of N- β -(aminoethyl)- γ -aminopropyltrimethoxysilane, 1 part of diphenyl silanediol, 2 parts of #918TM, and 5 parts of 2,4,6-tris(dimethylaminomethyl)phenol. These components were well mixed with each other in a nitrogen atmosphere in such a way to prevent contamination with moisture in air.

The test piece was prepared from the above composition following the above-described method of preparing the test piece, and cured at 23°C for 1 day and 50°C for another 3 days, to measure its adhesion strength. It had a tensile shear strength of 126 kg/cm² and a T-type releasing strength of 9.0 kg/25 mm (EXAMPLE 09).

<EXAMPLES P Series>

PRODUCTION EXAMPLE 1

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades

(agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber in a form of uniform solution.

10 A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

15 The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber thus produced contained ethylene at 68% by mol, and had an iodine value of 10, an intrinsic viscosity $[\eta]$ of 0.2dl/g, and M_w/M_n of 15.

20 Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of

the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber containing dimethoxymethylsilyl group.

EXAMPLES P1 and P2, and COMPARATIVE EXAMPLES P1 to P3

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A mixture containing the copolymer rubber containing
5 dimethoxysilyl group, prepared in PRODUCTION EXAMPLE P1, was prepared for each of EXAMPLES P1 and P2, and COMPARATIVE EXAMPLES P1 to P3. It was composed of the copolymer rubber, a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), colloidal calcium carbonate (Shiraishi K.K.,
10 EDS-D10ATM), limestone powder (Maruo Calcium, Snowlite SS TM), talc (Fuji Talc Kogyo, Talc LMRTM), Na₂SO₄•10H₂O, (Wako Jun-yaku Kogyo), and γ -isocyanate propyltriethoxysilane as a silane coupling agent (Nippon Unicar, Y-9030TM). Table P1 gives each composition in parts by weight. The mixture was well kneaded
15 by a 3-paint roll unit, to produce the major ingredient for each of the examples.

Each of the major ingredients was evaluated for its thready property by a spatula.

The major ingredient was incorporated with dibutyl tin
20 bisacetylacetonate (NITTO KASEI Co., Neostann U-220TM) to prepare the mixture of 404/2 ratio by weight. The components were well kneaded manually, and the resultant mixture was formed into a sheet in a 2mm thick aluminum frame lined with a Teflon sheet while breaking the bubbles in the composition by a spatula.

It was cured in an oven at 23°C for 7 days and 50°C for 7 days. The cured sheet was stamped out into the No.3 dumbbell-shaped test piece in accordance with JIS K-6251/1993, the "method of tensile testing vulcanized rubber," where the test piece was
5 stretched at 500mm/minute by an autograph (Shimadzu, Autograph AG-2000A) in a constant-temperature chamber kept at 23°C and 50±10%RH.

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The cured test piece for hardness measurement was prepared in the following procedure. The major ingredient was
10 incorporated with a curing catalyst (NITTO KASEI, Neostann U-220™) to prepare the mixture of 404/2 ratio by weight. The components were well kneaded, and the resultant mixture was cured into a rectangular parallelepiped test piece in a frame (12 by 12 by 50mm in size) lined with a Teflon sheet under the standard
15 conditions of 23°C for 7 days and 50°C also for 7 days, to measure its hardness. Hardness of the rod-shaped test piece was measured in accordance with JIS K-6301/1975 by the spring type hardness test A method using a rubber type hardness meter (Shimadzu, Hardness meter 200). A total of 5 measurements were made for
20 each composition, and the average value was reported.

Table P1 gives the composition, viscosity (at 10rpm) of the major ingredient, and thready property, dumbbell tensile test results, hardness, curing speed and resistance to weather of each composition.

Thready property was evaluated according to the following two-grade system:

○ : The composition is low in thready property, and easy to finish by spatula

5 × : The composition is high in thready property, and difficult to finish by spatula

M50, T_{\max} and E_{\max} in Table P1 are 50% tensile stress, maximum tensile stress and elongation at the maximum load, respectively.

10 Curing speed and resistance to weather were measured by the following methods:

1) Curing speed

The composition comprising the major ingredient and the catalyst was measured for its film expandability at room temperature, i.e., curing speed.

15 The curable composition was cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm.

(Evaluation standards)

20 × : when thickness of the cured portion is less than 1mm

○ : when thickness of the cured portion is 1mm or more

2) Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753, to determine resistance to weather:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

5 Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

The surface state of tested test piece was visually observed, to evaluate its resistance to weather according to the following three grades:

- 10 ○ : No cracks or molten portion observed
 △ : Cracks or molten portion observed slightly
 × : Cracks or molten portion observed

The results are given in Table P1.

COMPARATIVE EXAMPLES P4 to P8

- 15 An isobutylene polymer having a reactive silicon group was synthesized in accordance with the method described in Japanese Patent Laid-Open Publication No.316804/1998, paragraphs 0049 to 0055.

- 20 The composition was prepared for each of COMPARATIVE EXAMPLES P4 to P8 in the same manner as in each of EXAMPLES P1 and P2, and COMPARATIVE EXAMPLES P1 to P3, respectively, except that the copolymer rubber containing dimethoxysilyl group was replaced by the isobutylene polymer having a reactive silicon group. Several properties of the compositions were measured.

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The results are shown in Table P2

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Table P1

Compositions	Additives	EXAMPLE P		COMPARATIVE EXAMPLE P		
		1	2	1	2	3
Major ingredients (parts by weight)	Polymer produced in PRODUCTION EXAMPLE	100	100	100	100	100
	PS-32	100	100	100	100	100
	EDS-D10A	100	50	200	-	-
	Snowlite SS	50	200	-	300	-
	LMR	100	100	-	-	200
	Na ₂ SO ₄ ·10H ₂ O	2	2	2	2	2
Curing catalyst (parts by weight)	Y-9030	2	2	2	2	2
Thready property	U-220	2	2	2	2	2
		○	○	○	×	×
Tensile characteristics	M50 (kgf/cm ²)	7.90	7.91	3.52	4.12	9.69
	Tmax (kgf/cm ²)	11.7	12.6	11.6	8.8	13.1
	Emax (%)	85	83	215	118	62
JIS Hardness A		34	34	24	28	38
Curing speed		○	○	○	○	○
Resistance to weather		○	○	○	○	○

Table P2

Compositions	Additives	COMPARATIVE EXAMPLE P					
		4	5	6	7	8	
Major ingredients (parts by weight)	Note (1)	100	100	100	100	100	
	PS-32	100	100	100	100	100	
	EDS-D10A	200	-	-	100	50	
	Snowlite SS	-	300	-	50	200	
	LMR	-	-	200	100	100	
	Na ₂ SO ₄ •10H ₂ O	2	2	2	2	2	
Curing catalyst (parts by weight)	Y-9030	2	2	2	2	2	
	U-220	2	2	2	2	2	
Thready property		○	×	×	○	○	
Tensile characteristics	M50 (kgf/cm ²)	4.1	4.8	11.3	9.0	9.2	
	Tmax (kgf/cm ²)	13.8	10.4	15.4	13.8	14.7	
	E _{max} (%)	248	132	70	93	95	
JIS Hardness A Curing speed Resistance to weather		29	33	39	38	38	
		×	×	×	×	×	
		△	△	△	△	△	

Note (1): The polymer used in EXAMPLE described in Japanese Patent Laid-Open Publication No.316804/1998

It is apparent that the following findings are derived from the results given in Tables P1 and P2.

The composition prepared in each of EXAMPLES P1 and P2 using the silyl-containing copolymer rubber is excellent in
5 workability. The cured product is excellent in mechanical strength and hardness, and these properties are well-balanced. Moreover, it has sufficient resistance to weather and curing speed.

The composition prepared in each of COMPARATIVE EXAMPLES P1
10 to P3, which was free of calcium carbonate or talc although containing the silyl-containing copolymer rubber, was less balanced between these properties mentioned above.

The composition prepared in each of COMPARATIVE EXAMPLES P4
15 to P8, which was free of the silyl-containing copolymer rubber, was less balanced between these properties, and less resistant to weather or lower in curing speed.

<EXAMPLES Q Series>

The composition, the iodine value, the intrinsic viscosity
[η] and the molecular weight distribution (M_w/M_n) of the
20 copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined by the methods described earlier.

PRODUCTION EXAMPLE

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer

rubber (A-1)]

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The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber ($\text{A}_0\text{-1}$) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber ($\text{A}_0\text{-1}$) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g determined in decalin kept at 135°C , iodine value (IV) of 10(g/100g) and M_w/M_n of 15.

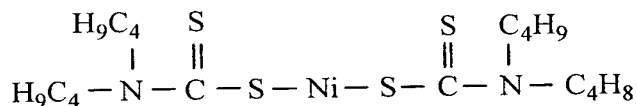
Two% toluene solution (0.3 part by weight) of chloroplatinic

acid and 1.5 parts by weight of methyldimethoxysilane were added to 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-Si(CH₃)(OCH₃)₂).

EXAMPLE Q1

A mixture containing the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE, was prepared for EXAMPLE Q1. It was composed of 100 parts of the copolymer rubber (A-1), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32™) as the plasticizer, 360 parts of limestone powder (Shiraishi Calcium, Whiton SB™) as the filler, 5 parts of salt cake Na₂SO₄•10H₂O as the water source, 1 part of tetrakis-[methylene-3-(3',5'-di-t-butyl-4-hydroxyphenyl)propionate]methane (Ciba-Geigy Japan, Irganox 1010™) as the aging inhibitor, 1 part of 2-(2'-hydroxy-3',5'-t-butyl)-5-chlorobenzotriazole

(Ciba-Geigy Japan, Tinuvin 327TM) as the stabilizer, 1 part of bis-(2,2,6,6-tetramethyl-4-piperidyl)cebacate (Sankyo, Sanol LS-770TM), 5 parts of a thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM), 2 parts of γ -isocyanate propyltriethoxysilane (Nippon Unicar, Y-9030TM) as the silane coupling agent, and 3 parts of nickel dimethyldithiocarbamate (Sanshin Kagaku Kogyo, Sandant NBCTM), represented by the following structural formula, as the light stabilizer, all parts by weight. It was sufficiently kneaded to mix the components by a 3-paint roll unit, to prepare the major ingredient.



The curing agent was prepared by the following procedure: a mixture comprising 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 25 parts of limestone powder (Shiraishi Calcium, Whiton SBTM), 4 parts of tetravalent tin compound (NITTO KASEI, U-220TM) as the curing catalyst, and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred repeatedly 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho, Excel Auto Homogenizer).

The test piece was prepared in accordance with JIS A-5758-1992

specifying the method of preparing the test piece for tensile
adhesion test; the composition comprising 14 parts by weight
of the above-described major ingredient and 1 part by weight
of the curing agent was put in the H-shape frame of glass or
5 aluminum substrate, after being sufficiently kneaded. The
composition was cured in an oven under the conditions of 23°C
for 7 days and 50°C for 7 days. The heat ray reflective glass
sheets (Central Glass, SGY-32TM and TCB-35TM, 5 by 5 by 0.6cm
in size) surface-coated with TiO₂ by sputtering were used as
10 the substrates for the H-shaped frames. Each H-shape frame was
washed with methylethylketone (Wako-Junyaku, special grade) and
wiped with clean cotton cloth, before it was filled with the
composition.

The light-resistant adhesion test was conducted using the
15 H-shaped test piece prepared as described above, wherein its
H-type mechanical properties before and after the test were
measured. The results are given in Table Q1. In the test, the
H-type tensile test piece, put in a Sunshine superlong-life
weathermeter (Suga Shikenki, WEL-SUN-HC) with black panel
20 temperature kept at 63°C, was irradiated with light emitted from
sunshine carbon as the light source, and taken out from the
analyzer after 480 hours.

The H-shape test piece thus prepared was tested by the method
of testing tensile adhesion in accordance with JIS A-5758/1992

at a tensile speed of 50mm/minute in a constant-temperature chamber kept at 23°C and 65±5%RH. The cohesion fracture (CF)/thin-coat fracture (TCF)/adhesion fracture (AF) ratio shown in the tables was determined by visual observation of the cross-sections of the tensile-tested pieces.

The cohesion fracture means the fracture of the cured composition itself, not at the interface between the base and cured composition, indicating that the cured composition is adhered to the base at a high adhesion strength. The adhesion fracture means the separation of the base and cured composition from each other at the interface, indicating that the cured composition is adhered to the base at a low adhesion strength. The thin-coat fracture is the cohesion fracture at the interface, indicating that its adhesion strength is medium between those for the above two fracture modes.

The weather resistance test was conducted by the following procedure. The results are also given in Table Q1.

Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions::

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

COMPARATIVE EXAMPLES Q1 to Q3

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The same procedures as used for EXAMPLE Q1 were repeated,
5 except that 3 parts by weight of nickel dimethyldithiocarbamate
(Sanshin Kagaku Kogyo, Sandant NBCTM) as the light stabilizer
was not incorporated (COMPARATIVE EXAMPLE Q1), 3 parts by weight
of 2-(2'-hydroxy-3',5'-t-butyl)-5-chlorobenzotriazole
(Ciba-Geigy Japan, Tinuvin 327TM) was incorporated as the
10 non-nickel-based stabilizer (COMPARATIVE EXAMPLE Q2), and 5
parts by weight of
bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate (Sankyo, Sanol
LS-770TM) was incorporated as the non-nickel-based stabilizer
(COMPARATIVE EXAMPLE Q3). The results are given in Table Q1.

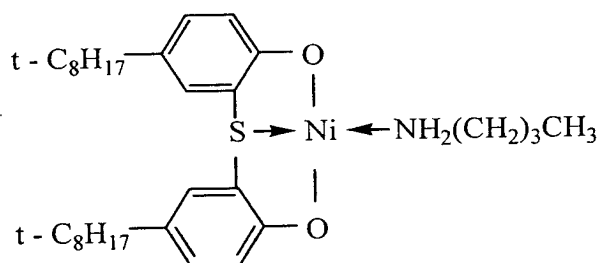
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Table Q1

Before Testing	M_{50} (kgf/cm ²)	T_{max} (kgf/cm ²)	E_{max} (%)	Fractured conditions (%)			Resistance to weather
				CF	TFC	AF	
EXAMPLE Q1	4.4	5.8	60	100	0	0	No cracks or molten portion observed
COMPARATIVE EXAMPLE Q1	4.3	5.9	62	99	1	0	No cracks or molten portion observed
COMPARATIVE EXAMPLE Q2	4.3	5.8	63	100	0	0	No cracks or molten portion observed
COMPARATIVE EXAMPLE Q3	4.3	5.6	60	100	0	0	No cracks or molten portion observed
After Testing	M_{50} (kgf/cm ²)	T_{max} (kgf/cm ²)	E_{max} (%)	Fractured conditions (%)			
				CF	TFC	AF	
EXAMPLE Q1	3.2	4.7	75	100	0	0	
COMPARATIVE EXAMPLE Q1	3.0	3.2	52	0	2	98	
COMPARATIVE EXAMPLE Q2	2.9	3.4	54	0	1	99	
COMPARATIVE EXAMPLE Q3	3.1	3.1	50	1	0	99	

As shown in Table Q1, the composition prepared in each of
 EXAMPLE Q1 and COMPARATIVE EXAMPLES Q1 to Q3 exhibited good
 adhesion to the surface-treated SGY-32 glass substrate before
 the weather-resistant adhesion test. After the test, on the
 5 other hand, only the test piece prepared in EXAMPLE Q1 showed
 the cohesion fracture, and the others the adhesion fracture.
 These results indicate that incorporation of the Ni-based
 stabilizer (Sandant NBC™) improves light-resistant adhesion.
EXAMPLE Q2 and COMPARATIVE EXAMPLE Q4

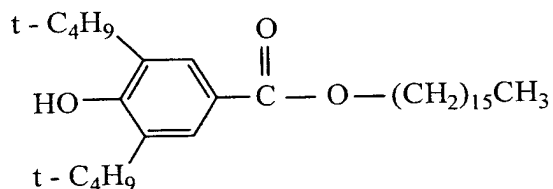
10 The same procedures as used for EXAMPLE Q1 were repeated for
 EXAMPLE Q2, except that 3 parts by weight of nickel
 dimethyldithiocarbamate (Sanshin Kagaku Kogyo, Sandant NBC™)
 was replaced by 3 parts by weight of nickel [2,2'-
 -thiobis(4-t-octylphenolate)]-n-butylamine (ACC, SYASORB
 15 UV1084), represented by the following formula, also as the
 Ni-based light stabilizer:



The same procedures as used for EXAMPLE Q2 were repeated for
 COMPARATIVE EXAMPLE Q4, except that 3 parts by weight of nickel

[2,2'-thiobis(4-t-octylphenolate)]-n-butylamine (ACC, SYASORB UV1084TM) was replaced by 3 parts by weight of 3,5-di-t-butyl-4-hydroxybenzoic acid-n-hexadecyl ester (ACC, SYASORB UV2908TM), represented by the following formula, as the

5 non-Ni-based light stabilizer:



10

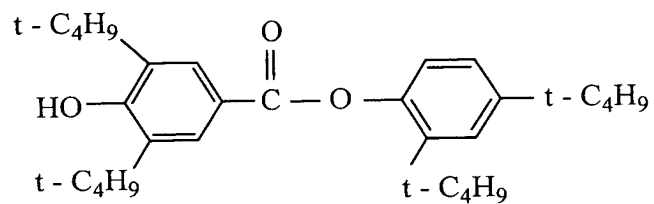
The results are given in Table Q2.

EXAMPLE Q3 and COMPARATIVE EXAMPLE Q5

The same procedures as used for EXAMPLE Q1 were repeated for EXAMPLE Q3, except that quantity of limestone powder (Shiraishi Calcium, Whiton SBTM) was reduced from 25 to 20 parts by weight.

15 The same procedures as used for EXAMPLE Q3 were repeated for COMPARATIVE EXAMPLE Q5, except that 3 parts by weight of nickel dimethyldithiocarbamate (Sanshin Kagaku Kogyo, Sandant NBCTM) as the Ni-based light stabilizer was replaced by 3 parts by weight

20 of 2,4-dibutylphenyl-3',5'-di-t-butyl-4'-hydroxybenzoate (ACC, SYASORB 712TM), represented by the following formula, as the non-Ni-based light stabilizer:



5

The results are given in Table Q2.

10

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Table Q2

	M_{50} (kgf/cm ²)	T_{max} (kgf/cm ²)	E_{max} (%)	Fractured conditions (%)			Resistance to weather
				CF	TFC	AF	
EXAMPLE Q2	3.2	4.4	78	100	0	0	No cracks or molten portion observed
EXAMPLE Q3	3.3	4.8	80	100	0	0	No cracks or molten portion observed
COMPARATIVE EXAMPLE Q4	3.0	3.2	52	0	1	99	No cracks or molten portion observed
COMPARATIVE EXAMPLE Q5	3.2	3.2	50	0	2	98	No cracks or molten portion observed

<EXAMPLES R Series>

The composition, the iodine value, the intrinsic viscosity [η] and the molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE

5 EXAMPLES were determined by the methods described earlier.

Curing speed and resistance to weather tests were conducted in EXAMPLES and COMPARATIVE EXAMPLES by the following methods:

(1) Curing speed test

The compositions prepared in EXAMPLES and COMPARATIVE

10 EXAMPLES, were cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and then released from the mold. Thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was more than 1mm, △ when

15 it was 0.5 to 1mm, and × when it was less than 0.5mm

(2) Weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 using a Sunshine Carbon Arc weatherometer, to determine resistance to weather:

20 <Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: 40±2°C

Total light irradiation time: 500 hours

<Evaluation standards for resistance to weather>

○ : No cracks or molten portion observed on one side of the tested piece

5 △ : Cracks or molten portion observed slightly on one side of the tested piece

× : Cracks or molten portion observed on one side of the tested piece

PRODUCTION EXAMPLE R1

10 [Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

15 The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ 20 and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity [η] of 0.2dl/g determined in decalin kept at 135°C, iodine value (IV) of 10(g/100g) and Mw/Mn of 15.

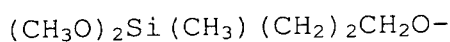
Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-SiCH₃(OCH₃)₂).

PRODUCTION EXAMPLE R2

Two hundred grams of a polypropylene oxide with allyl ether group at 97% of the total terminals and having an average molecular weight of 5,000, was charged in a 500mL four-mouthed flask, to

which 100 μ L of a 10% ethanol solution of chloroplatinic acid was added and then methyldimethoxysilane was added dropwise at 50°C, and they were allowed to react with each other at 80°C for around 3 hours.

- 5 The resultant reaction product was a polypropylene oxide having 1.7 structures represented by the following formula in one molecule and an average molecular weight of around 5,200, as revealed by the ^1H -NMR analysis:



10 EXAMPLES R1 to R6

- The silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared by PRODUCTION EXAMPLE R1, was used to prepare the toluene solution of uniform composition, described in Table R1, for each of EXAMPLES R1 to R6.

Each solution was cured at room temperature for 1 day and at 50°C for another 4 days in an about 3mm thick frame, and then treated at 50°C for 2 hours under a vacuum of 2 to 3mmHg, to completely evaporate toluene.

- 20 The resultant cured sheet was put in a hot wind type drier kept at 150°C for 20 days, to observe temporal property changes (measurement of resistance to heat). The results are given in Table R1, wherein the resistance to heat was evaluated by the three grades, O: no cracks or molten portion observed, Δ : cracks

- or molten portion observed slightly, and *: cracks or molten
portion observed; and NISSAN DLTP: sulfide carboxylate
ester-based antioxidant (NOF Corp.), Nocrac 300:
sulfur-containing hindered phenol (Ouchishinko Chemical
5 Industries Co.,) and Irgano 1010: hindered phenol (Ciba Geigy,
Japan) .

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Table R1

EXAMPLES	Compositions [parts by weight]							Properties		
	Component (A1)	Component (U)		Tin octylate	Lauryl amine	Toluene	Water	Resistance to heat	Resistance to weather	Curing speed
R1	100	NISSAN DLTP	1	3	0.75	50	0.5	○	○	○
R2	100	NISSAN DLTP	3	3	0.75	50	0.5	○	○	○
R3	100	NISSAN DLTP	5	3	0.75	50	0.5	○	○	○
R4	100	Nocrac 300	1	3	0.75	50	0.5	○	○	○
R5	100	Nocrac 300	3	3	0.75	50	0.5	○	○	○
R6	100	Nocrac 300	5	3	0.75	50	0.5	○	○	○

Component (A1): ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)

EXAMPLES R7 to R9, and COMPARATIVE EXAMPLE R1

The 1mm thick sheet was prepared for each of EXAMPLES R7 to R9, and COMPARATIVE EXAMPLE R1 in the same manner as in EXAMPLE R1, except that the sulfur-based aging inhibitor was replaced
5 by the additive given in Table R2 to prepare the composition. A test tube type rubber aging tester was used to measure the time required for the sheet to be completely decomposed at 150°C and start to flow.

The results are given in Table R2.

10 COMPARATIVE EXAMPLE R2

The sheet was prepared for COMPARATIVE EXAMPLE R2 in the same manner as in EXAMPLE R7, except that the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) prepared in PRODUCTION EXAMPLE R1 was replaced by
15 the polymer prepared in PRODUCTION EXAMPLE R2, and tested also in the same manner. The results are given in Table R2.

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Table R2

	EXAMPLE R			COMPARATIVE EXAMPLE R	
	7	8	9	1	2
NISSAN DLTP [parts by weight]	1	-	1	-	1
Nocrac 300 [parts by weight]	-	1	1	-	-
Resistance to heat	○	○	○	△	△
Resistance to weather	○	○	○	△	×
Curing speed	○	○	○	○	○

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The cured composition prepared in each of EXAMPLES R1 to R9 and COMPARATIVE EXAMPLES R1 and R2 was tested for resistance to weather following the method described earlier. As a result, no cracks were observed in the test pieces prepared in EXAMPLES R1 to R9, but observed in those prepared in COMPARATIVE EXAMPLES R1 and R2.

<EXAMPLES S Series>

The composition, the iodine value, the intrinsic viscosity $[\eta]$ and the molecular weight distribution (M_w/M_n) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined by the methods described earlier.

PRODUCTION EXAMPLE

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $VO(OEt)_2Cl$ and $Al(Et)_{1.5}Cl_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an intrinsic viscosity $[\eta]$ of 0.2dl/g determined in decalin kept at 135°C, iodine value (IV) of 10g/100g and Mw/Mn of 15.

Two% toluene solution (0.3 part by weight) of chloroplatinic acid and 1.5 parts by weight of methyldimethoxysilane were added to 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group $(-\text{Si}(\text{CH}_3)(\text{OCH}_3)_2)$.

EXAMPLES S1 to S5

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A mixture containing the polymer prepared in PRODUCTION
EXAMPLE was prepared for each of EXAMPLES S1 to S5. It was
composed of 100 parts of the polymer, 90 parts of a paraffin-based
5 process oil (Idemitsu Kosan, Diana Process Oil PS-32TM),
30 parts of limestone powder (Maruo Calcium, Snowlite SSTM),
100 parts of colloidal calcium carbonate (Shiraishi K.K.,
EDS-D10ATM), 100 parts of talc (Fuji Talc Kogyo, Talc LMRTM),
6 parts of Na₂SO₄·10H₂O, 6 parts of china wood oil as the compound
10 and component (V) for the present invention having an unsaturated
group polymerizable on reacting with oxygen in air, 3 parts of
dipentaerythritol penta- and hexa-acrylate as the
photopolymerizable compounds (TOAGOSEI, Aronix M-400TM), and
a tackifier given in Table S1 in a quantity also given in Table
15 S1, all parts by weight. Each mixture was well kneaded by a
3-paint roll unit, to produce the major ingredient for each
example.

The tackifiers used in EXAMPLES S1 to S3 were γ
-glycidoxypropyltrimethoxysilane (Nippon Unicar, Silane
20 coupling agent A-187TM) as the silane coupling agent for the
present invention and/or γ -isocyanatepropyltriethoxysilane
(Nippon Unicar, Silane coupling agent Y-9030TM) in quantities
given in Table S1. EXAMPLE S4 used no tackifier, while EXAMPLE
S5 used 4 parts by weight of an epoxy resin as the tackifier

containing no hydrolyzable silicon group (except the silane coupling agent) (Yuka Shell Epoxy, Epikote #828TM).

The curing agent was prepared by the following procedure:
a mixture comprising 10 parts of a paraffin-based process oil
5 (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), 4 parts of a curing catalyst (NITTO KASEI, U-220TM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for
10 10 minutes by a homogenizer (Nihon Seiki Seisakusho Co., Ltd., Excel Auto Homogenizer).

The cured test piece for tensile adhesion measurement was prepared in the following procedure in accordance with JIS A-5758/1992. The major ingredient was incorporated with the
15 curing agent to prepare the mixture of 12/1 by weight. The components were well kneaded, and the resultant mixture was put in the H-shape of glass base while breaking the bubbles in the composition by a spatula, and cured in an oven, under the conditions of 23°C×1 day + 50°C×5 days for each composition.
20 The substrate for the H-type tensile test was of float glass (Koen-sha, designated by Japan Sealant Industry Association, 3 by 5 by 0.5cm in size) in accordance with JIS A-5758/1992, or heat ray reflective glass (Central Glass, KLSTM, 5 by 5 by 0.6cm in size) coated with thermally fused TiOx. Each of these

H-shaped bases was washed with methylethylketone (Wako-Junyaku, special grade) and wiped with clean cotton cloth, before it was filled with the composition. It was not coated with a primer.

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The H-shape test piece thus prepared for the tensile test
5 was tested after it was cured in the absence of primer by the method of testing tensile adhesion in accordance with JIS A-5758/1992. The test was conducted at a tensile speed of 50 mm/minute by an autograph (Shimadzu, Autograph AG-2000A) in a constant-temperature chamber kept at 23°C and 50±10%RH, to
10 evaluate adhesion in the absence of a primer by comparing the tensile characteristics with fractured morphology. The results are given in Table S1, where the cohesion fracture (CF)/thin-coat fracture (TCF)/adhesion fracture (AF) ratio was determined by visual observation of the cross-sections of each tensile-tested
15 piece.

Curing speed and resistance to weather were measured by the following methods. The results are given in Table S7.

(1) Curing speed test

The curable composition was cured at 23°C and 50%RH for 24
20 hours in a mold (20 by 80 by 5mm in size), and then released from the mold. Thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm, to evaluate its curing speed. It was marked with ○ when its thickness was 1mm or more, and × when it was less than 1mm.

(2) Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions, to determine resistance to weather:

5 Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^{\circ}\text{C}$

Tank inside temperature: $40 \pm 2^{\circ}\text{C}$

10 Total light irradiation time: 1000 hours

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*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%
*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

EXAMPLES S6 to S9, and COMPARATIVE EXAMPLE S1

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A mixture containing the polymer prepared in PRODUCTION EXAMPLE was prepared for each of EXAMPLES S6 to S9, and COMPARATIVE EXAMPLE S1. It was composed of 100 parts of the polymer, 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 30 parts of limestone powder (Maruo Calcium, Snowlite SSTM), 100 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Fuji Talc Kogyo, Talc LMRTM), 6 parts of Na₂SO₄·10H₂O, 3 parts of nickel dimethyldithiocarbamate (Sanshin Kagaku Kogyo, Sandant NBCTM) as a light stabilizer, 3 parts of nickel [2,2'-thiobis(4-t-octylphenolate)]-n-butylamine (ACC, CYASORB UV-1084), 1 part of antioxidant (Ciba-Geigy Japan, Irganox 1010TM), 1 part of ultraviolet ray absorber (Ciba-Geigy Japan, Tinuvin 327TM), 1 part of light stabilizer (Sankyo, Sanol LS-770TM), and 2 parts of γ -glycidoxypropyltrimethoxysilane (Nippon Unicar, Silane coupling agent A-187TM) as the silane coupling agent for the present invention and 4 parts of γ -isocyanatepropyltriethoxysilane (Nippon Unicar, Silane coupling agent Y-9030TM), all parts by weight. It was also incorporated with china wood oil as the compound and component (V) for the present invention having an unsaturated group polymerizable on reacting oxygen in air, and dipentaerythritol penta- and hexa-acrylate as the photopolymerizable compounds

(TOAGOSEI, Aronix M-400TM) in a quantity given in Table S2. Each mixture was well kneaded by a 3-roll unit, to produce the major ingredient for each example.

The curing agent was prepared by the following procedure:

5 a mixture comprising 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SS), 4 parts of a curing catalyst (NITTO KASEI, U-220TM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded
10 in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd., Excel Auto Homogenizer).

The H-shaped test piece was prepared in the same manner as in the above, except that it was cured under the conditions of
15 23°C×7 days + 50°C×7 days. The H-type tensile test piece, put in a Sunshine superlong-life weathermeter (Suga Shikenki, WEL-SUN-HC) with black panel temperature kept at 63°C, was exposed to light emitted from sunshine carbon as the light source in the weather resistance tester (SWOM) for a time given in Table
20 S2, and taken out from the tester to be tested for tensile adhesion. Its weather-resistant adhesion was evaluated by comparing the tensile characteristics with fractured morphology. The results are given in Table S2.

The H-shaped test piece was also tested for tensile adhesion

for reference before it was tested for weather-resistance. The results are given in Table S3.

The composition prepared in each of EXAMPLES S6 to S9 was tested for curing speed and resistance to weather in the same
5 manner as in EXAMPLES S1 to S3. The results are also given in Table S7.

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Table S2

	Unsaturated compound capable of reacting with oxygen in air (Addition rate)	Photopolymerizable Material (Addition rate)	Base materials	SWOM Exposure time	Elongation at max. load *1	Fractured morphologies (%) *2		
						CF	TCF	AF
COMPARATIVE EXAMPLE S1	Not used	Not used	Float glass	3000	×	0	0	100
			Heat ray reflective glass:KLS	500	×	100	0	0
EXAMPLE S6	Not used	AronixM400 (3 parts)	Float glass	3000	×	50	50	0
			Heat ray reflective glass:KLS	600	×	0	0	100
EXAMPLE S7	Not used	AronixM400 (6 parts)	Heat ray reflective glass:KLS	500	○	100	0	0
			Float glass	3000	○	100	0	0
EXAMPLE S8	China wood oil (6 parts)	Not used	Heat ray reflective glass:KLS	1000	○	100	0	0
			Float glass	3000	○	100	0	0
EXAMPLE S9	China wood oil (6 parts)	AronixM400 (3 parts)	Float glass	3000	○	100	0	0
			Heat ray reflective glass:KLS	1000	○	100	0	0

*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%

*2 CF: Cohesion fracture, TCF: Thin-coated fracture, AF: Adhesion fracture

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Table S3

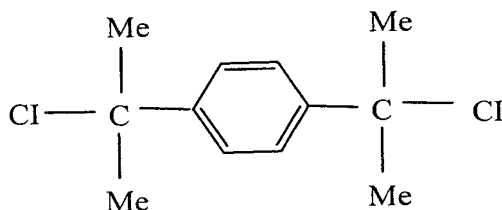
	Unsaturated compound capable of reacting with oxygen in air (Addition rate)	Photopolymerizable Material (Addition rate)	Base materials	Elongation at max. load *1	Fractured morphologies (%) *2		
					CF	TCF	AF
COMPARATIVE EXAMPLE S1	Not used	Not used	Float glass	△	100	0	0
			Heat ray reflective glass:KLS	△	100	0	0
EXAMPLE S6	Not used	AronixM400 (3 parts)	Float glass	△	100	0	0
			Heat ray reflective glass:KLS	○	100	0	0
EXAMPLE S7	Not used	AronixM400 (6 parts)	Heat ray reflective glass:KLS	△	100	0	0
EXAMPLE S8	China wood oil(6 parts)	Not used	Float glass	○	100	0	0
			Heat ray reflective glass:KLS	△	100	0	0
EXAMPLE S9	China wood oil(6 parts)	AronixM400 (3 parts)	Float glass	○	100	0	0
			Heat ray reflective glass:KLS	○	100	0	0

*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%

*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

REFERENCE PRODUCTION EXAMPLE

A 500mL pressure-resistant glass reactor was charged, after it was equipped with a 3-way cock and purged inside with nitrogen, 54mL of ethyl cyclohexane (dried by molecular sieves 3A at least for a night), 126mL of toluene (also dried by molecular sieves 3A at least for a night) and 1.16g (5.02mmols) of p-DCC represented by the following formula by a syringe.



Next, a pressure-resistant glass-made liquefied gas collecting tube equipped with a needle valve and containing 56mL of isobutylene monomer was connected to the 3-way cock. Then the reactor for polymerization was immersed in a dry ice/ethanol bath kept at -70°C to cool the solution, and evacuated to a vacuum. It was then charged with isobutylene monomer from the liquefied gas collecting tube by opening the needle valve, and returned back to the normal pressure through introducing a nitrogen gas by handling the 3-way cock. The reactor was charged with 0.093g (1.0mmol) of 2-methylpyridine and then with 1.65mL (15.1mmols) of titanium tetrachloride, to initiate the polymerization. After a lapse of 70 minutes, 1.22g (10.8mmols) of allyl

trimethylsilane was added to the reactor to introduce the allyl group at the polymer terminal. After a lapse of 120 minutes for the reaction, the reaction solution was washed 4 times each with 200mL of water, and the solvent was distilled off to obtain

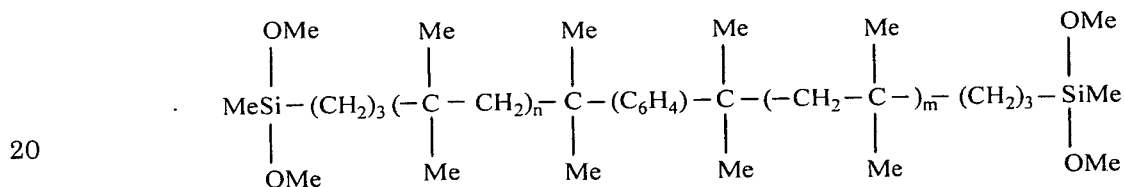
5 the isobutylene-based polymer with the allyl group at the terminal.

Next, 40g of the isobutylene-based polymer with the allyl group at the terminal thus obtained was dissolved in 20mL of n-heptane, and the mixture was heated to around 70°C, to which

10 1.5 [eq/vinyl group] of methyl dimethoxysilane and 1×10^{-4} [eq/vinyl group] of a platinum/vinyl siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 4 hours.

The reaction solution was concentrated under a vacuum, to

15 produce the isobutylene polymer with the reactive silicon groups at both terminals, represented by the following formula:



The polymer yield was estimated from the quantity produced. It was also analyzed for Mn and Mw/Mn by GPC, and the terminal

structure by comparing the intensities of the 300MHz
 ^1H -NMR-analyzed resonance signals of proton relevant to each
structure (proton derived from the initiator: 6.5 to 7.5ppm,
methylprotonbondedtothesiliconatom, derivedfromthepolymer
5 terminal: 0.0 to 0.1ppm, and methoxy proton: 3.4 to 3.5ppm) with
each other.

The ^1H -NMR analysis was conducted using a Varian Gemini 300
(300MHz for ^1H) in CDCl_3 .

10 The FT-IR analysis was conducted by an IR analyzer (Shimadzu
IR-408), and GPC analysis was conducted with a Waters LC Module
1 as the liquid sending system and Shodex K-804 as the column.
The molecular weight was the one relative to the polystyrene
standard. The polymer thus prepared had an Mn of 11,400, Mw/Mn
of 1.23 and Fn (silyl) of 1.76, wherein the number-average
15 molecular weight was as polystyrene, and the number of the
terminal silyl functional group was that per 1 mol of isobutylene
polymer.

REFERENCE EXAMPLES S1 to S5

20 A mixture containing the polymer prepared in REFERENCE
PRODUCTION EXAMPLE was prepared for each of REFERENCE EXAMPLES
S1 to S5. It was composed of 100 parts of the polymer, 90 parts
of a paraffin-based process oil (Idemitsu Kosan, Diana Process
Oil PS-32TM), 30 parts of limestone powder (Maruo Calcium,
Snowlite SSTM), 100 parts of colloidal calcium carbonate

(Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Fuji Talc Kogyo, Talc LMRTM), 6 parts of Na₂SO₄•10H₂O, 6 parts of china wood oil as the compound and component (V) for the present invention having an unsaturated group polymerizable on reacting with oxygen in
5 air, 3 parts of dipentaerythritol penta- and hexa-acrylate as the photopolymerizable compounds (TOAGOSEI, Aronix M-400TM), and a tackifier given in Table S4 in a quantity also given in Table S4, all parts by weight. Each mixture was well kneaded by a 3-paint roll unit, to produce the major ingredient for each
10 example.

The tackifiers used in REFERENCE EXAMPLES S3 to S5 were γ -glycidoxypropyltrimethoxysilane (Nippon Unicar, Silane coupling agent A-187TM) as the silane coupling agent and/or γ -isocyanatepropyltriethoxysilane (Nippon Unicar, Silane
15 coupling agent Y-9030TM) in quantities given in Table S1. REFERENCE EXAMPLE S1 used no tackifier, while REFERENCE EXAMPLE S2 used 4 parts by weight of an epoxy resin as the tackifier containing no hydrolyzable silicon group (except the silane coupling agent) (Yuka Shell Epoxy, Epikote #828TM).

20 The curing agent was prepared by the following procedure: a mixture comprising 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), 4 parts of a curing catalyst (NITTO KASEI, U-220TM), and 2.5 parts of carbon black (Mitsubishi

Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd., Excel Auto Homogenizer).

5 The test piece was prepared in accordance with JIS A-5758/1992 for the tensile adhesion test; 12 parts by weight of the major ingredient and 1 part by weight of the curing agent were well kneaded, and the resultant mixture was put in the H-shape of glass base while breaking the bubbles in the composition by a
10 spatula and cured in an oven under the conditions of 23°C×1 day + 50°C×5 days for each composition. The substrate for the H-type tensile test was of float glass (Koen-sha, designated by Japan Sealant Industry Association, 3 by 5 by 0.5cm in size) in accordance with JIS A-5758/1992, or heat ray reflective glass
15 (Central Glass, KLSTM, 5 by 5 by 0.6cm in size) coated with thermally fused TiO_x. Each of these H-shaped bases was washed with methylethylketone (Wako-Junyaku, special grade) and wiped with clean cotton cloth, before it was filled with the composition. It was not coated with a primer.

20 The H-shape test piece thus prepared for the tensile test was tested after it was cured in the absence of primer by the method of testing tensile adhesion in accordance with JIS A-5758/1992. The test was conducted at a tensile speed of 50mm/minute by an autograph (Shimadzu, Autograph AG-2000A) in

a constant-temperature chamber kept at 23°C and 50±10%RH, to evaluate adhesion in the absence of a primer by comparing the tensile characteristics with fractured morphology. The results are given in Table S4, where the cohesion fracture (CF)/thin-coat fracture (TCF)/adhesion fracture (AF) ratio was determined by visual observation of the cross-sections of each tensile-tested piece.

Curing speed and resistance to weather were measured in REFERENCE EXAMPLES S3 to S5 in the same manner as in EXAMPLES S1 to S3. The results are given in Table S7.

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Table S4

	Tackifier (Addition rate)	Base materials	Elongation at max. load * ₁	Fractured morphologies (%) * ₂		
				CF	TCF	AF
REFERENCE EXAMPLE S1	Not used	Float glass	×	0	0	100
		Heat ray reflective glass:KLS	×	0	0	100
REFERENCE EXAMPLE S2	Epikote 828 (4 parts)	Float glass	×	0	0	100
		Heat ray reflective glass:KLS	×	0	0	100
REFERENCE EXAMPLE S3	A-187 (4 parts)	Float glass	○	100	0	0
		Heat ray reflective glass:KLS	○	100	0	0
REFERENCE EXAMPLE S4	Y-9030 (4 parts)	Float glass	△	100	0	0
		Heat ray reflective glass:KLS	△	100	0	0
REFERENCE EXAMPLE S5	A-187 (2 parts) Y-9030 (4 parts)	Float glass	○	100	0	0
		Heat ray reflective glass:KLS	△	100	0	0

*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%

*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

REFERENCE EXAMPLES S6 to S10

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A mixture containing the polymer prepared in REFERENCE PRODUCTION EXAMPLE was prepared for each of REFERENCE EXAMPLES S6 to S10 in the same manner as in EXAMPLES S6 to S9 and COMPARATIVE EXAMPLE S1. It was composed of 100 parts of the polymer, 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 30 parts of limestone powder (Maruo Calcium, Snowlite SSTM), 100 parts of colloidal calcium carbonate (Shiraishi K.K., EDS-D10ATM), 100 parts of talc (Fuji Talc Kogyo, Talc LMRTM), 6 parts of Na₂SO₄•10H₂O, 3 parts of nickel dimethyldithiocarbamate (Sanshin Kagaku Kogyo, Sandant NBCTM) as the light stabilizer, 3 parts of nickel [2,2'-thiobis(4-t-octylphenolate)]-n-butylamine (ACC, CYASORB UV-1084), 1 part of antioxidant (Ciba-Geigy Japan, Irganox 1010TM), 1 part of ultraviolet ray absorber (Ciba-Geigy Japan, Tinuvin 327TM), 1 part of light stabilizer (Sankyo, Sanol LS-770TM), and 2 parts of γ -glycidoxypropyltrimethoxysilane (Nippon Unicar, Silane coupling agent A-187TM) as the silane coupling agent and 4 parts of γ -isocyanatepropyltriethoxysilane (Nippon Unicar, Silane coupling agent Y-9030TM), all parts by weight. It was also incorporated with china wood oil as the compound and component (V) for the present invention having an unsaturated group polymerizable on reacting with oxygen in air, and

dipentaerythritol penta- and hexa-acrylate as the photopolymerizable compounds (TOAGOSEI, Aronix M-400TM) in a quantity given in Table S5. Each mixture was well kneaded by a 3-roll unit, to produce the major ingredient for each example.

The curing agent was prepared by the following procedure: a mixture comprising 10 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 20 parts of limestone powder (Maruo Calcium, Snowlite SSTM), 4 parts of a curing catalyst (NITTO KASEI, U-220TM), and 2.5 parts of carbon black (Mitsubishi Chemical, CB#30TM), all parts by weight, was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd., Excel Auto Homogenizer).

The H-shaped test piece was prepared in the same manner as in the above, except that it was cured under the conditions of 23°C×7 days + 50°C×7 days. The H-type tensile test piece, put in a Sunshine superlong-life weathermeter (Suga Shikenki, WEL-SUN-HC) with black panel temperature kept at 63°C, was exposed to light emitted from sunshine carbon as the light source in the weather resistance tester (SWOM) for a time given in Table S5, and taken out from the tester to be tested for tensile adhesion. Its weather-resistant adhesion was evaluated by comparing the tensile characteristics with fractured morphology. The results

are given in Table S5.

The H-shaped test piece prepared as described above was also tested for tensile adhesion for reference before it was tested for weather-resistance. The results are also given in Table

5 S6.

The composition prepared in each of REFERENCE EXAMPLES S7 to S10 was tested for curing speed and resistance to weather in the same manner as in EXAMPLES S1 to S3. The results are also given in Table S7.

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Table S5

	Unsaturated compound capable of reacting with oxygen in air (Addition rate)	Photopolymerizable Material (Addition rate)	Base materials	SWOM Exposure time	Elongation at max. load *1	Fractured morphologies (%) *2		
						CF	TCF	AF
REFERENCE EXAMPLE S6	Not used	Not used	Float glass Heat ray reflective glass:KLS	3000 500	△ △	100 0	0 0	0 100
REFERENCE EXAMPLE S7	Not used	AronixM400 (3 parts)	Float glass Heat ray reflective glass:KLS	3000 600	△ △	100 70	0 0	0 30
REFERENCE EXAMPLE S8	Not used	AronixM400 (6 parts)	Heat ray reflective glass:KLS	500	×	50	0	50
REFERENCE EXAMPLE S9	China wood oil(6 parts)	Not used	Float glass Heat ray reflective glass:KLS	3000 1000	△ △	100 100	0 0	0 0
REFERENCE EXAMPLE S10	China wood oil(6 parts)	AronixM400 (3 parts)	Float glass Heat ray reflective glass:KLS	3000 1000	△ △	100 100	0 0	0 0

*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%

*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

Table S6

	Unsaturated compound capable of reacting with oxygen in air (Addition rate)	Photopolymerizable Material (Addition rate)	Base materials	Elongation at max. load *1	Fractured morphologies (%) *2		
					CF	TCF	AF
REFERENCE EXAMPLE S6	Not used	Not used	Float glass Heat ray reflective glass:KLS	△	97	3	0
REFERENCE EXAMPLE S7	Not used	AronixM400 (3 parts)	Float glass Heat ray reflective glass:KLS	△	98	2	0
REFERENCE EXAMPLE S8	Not used	AronixM400 (6 parts)	Heat ray reflective glass:KLS	×	98	2	0
REFERENCE EXAMPLE S9	China wood oil (6 parts)	Not used	Float glass Heat ray reflective glass:KLS	△	100	0	0
REFERENCE EXAMPLE S10	China wood oil (6 parts)	AronixM400 (3 parts)	Float glass Heat ray reflective glass:KLS	×	100	0	0

*1 ○: Elongation of 80% or more, △: Elongation of less than 80%, ×: Elongation of less than 60%

*2 CF: Cohesion fracture, TCF: Thin-coat fracture, AF: Adhesion fracture

Table S7

	Curing speed*	Resistance to weather
EXAMPLE S1	○	No cracks or molten portion observed
EXAMPLE S2	○	No cracks or molten portion observed
EXAMPLE S3	○	No cracks or molten portion observed
EXAMPLE S6	○	No cracks or molten portion observed
EXAMPLE S7	○	No cracks or molten portion observed
EXAMPLE S8	○	No cracks or molten portion observed
EXAMPLE S9	○	No cracks or molten portion observed
REFERENCE EXAMPLE S3	×	Cracks or molten portion observed
REFERENCE EXAMPLE S4	×	Cracks or molten portion observed
REFERENCE EXAMPLE S5	×	Cracks or molten portion observed
REFERENCE EXAMPLE S7	×	Cracks or molten portion observed
REFERENCE EXAMPLE S8	×	Cracks or molten portion observed
REFERENCE EXAMPLE S9	×	Cracks or molten portion observed
REFERENCE EXAMPLE S10	×	Cracks or molten portion observed

* ○: Sufficiently serviceable, △: Tackiness remaining, ×: Uncured

<EXAMPLES T Series>

The composition, the iodine value, the intrinsic viscosity $[\eta]$ and the molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE

5 EXAMPLES were determined by the methods described earlier.

Release-resisting force to silicone release paper, residual tackiness, tackiness, curing speed and weather resistance of the compositions prepared in EXAMPLES and COMPARATIVE EXAMPLES were determined by the following methods.

10 (1) Release-resisting force to silicone release paper

The adhesive tape was prepared and put on a commercially available silicone release paper, to prepare the test piece. It was kept at 50°C for 7, 14 or 21 days for the accelerated adhesion, taken out and returned back to the normal temperature,

15 and tested for its release-resisting force, defined as the resistance of the adhesive tape when it was released by 180° from the silicone release paper at a tensile speed of 300mm/minute.

(2) Residual tackiness

20 The adhesive tape was prepared and put on a commercially available silicone release paper, to prepare the test piece. It was left at 50°C, and the adhesive tape was released, to measure its tackiness. The residual tackiness is defined as the above tackiness relative to its initial tackiness, reported in

percentage.

(3) Tackiness

The adhesive tape was prepared and put on a stainless steel plate, to prepare the test piece. It was left at 23°C for 60 minutes, and tested for its tackiness, defined as the release strength of the adhesive tape when it was released by 180° from the stainless steel plate at a tensile speed of 300mm/minute at 23°C.

(4) Curing speed

The curable composition was cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size).

The cured composition was released from the mold, and thickness of the cured portion of the composition was measured by a dial gauge of weak spring force to 0.1mm. It was marked with ○ when its thickness was 1mm or more, and × when it was less than 1mm.

Moreover, the time required for the composition to be cured under the conditions of 120°C and 50%RH was measured. The composition was marked with ○ when it was cured in less than 5 minutes, △ when it was cured in 5 to 10 minutes, and × when it was cured in more than 10 minutes.

(5) Weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 using a Sunshine Carbon Arc weatherometer, to determine resistance to weather:

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<Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

5 Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

PRODUCTION EXAMPLE T1

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
10 rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene,
15 propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

20 The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate

the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber thus produced contained ethylene at 68% by mol, and had an iodine value of 10(g/100g), an intrinsic viscosity $[\eta]$, as measured in decalin kept at 135°C, of 0.2dl/g, and Mw/Mn of 15.

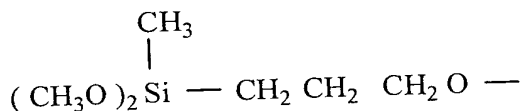
Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-SiCH₃(OCH₃)₂).

PRODUCTION EXAMPLE T2

Eight hundred grams of a polypropylene oxide (produced from polypropylene glycol as the starting material) with allyl ether group at 98% of the total terminals and having an average molecular weight of around 8,000, was charged in an agitator-equipped, pressure-resistant reactor, to which 20g of

methyldimethoxysilane was added, and then 0.34mL of a catalyst solution of chloroplatinic acid (8.9g of $\text{H}_2\text{PtCl}_6 \cdot 6\text{H}_2\text{O}$ dissolved in 18mL of isopropyl alcohol and 160mL of tetrahydrofuran) was added, and they were allowed to react with each other at 80°C for 6 hours.

The unreacted silane was quantitatively analyzed by gas chromatography and infrared analysis, to find the conversion rate. The results indicated that a polypropylene oxide with the following group at 84% of its total terminals was produced:



PRODUCTION EXAMPLE T3

Tetra-n-butoxy zirconium, 38.3g (0.1mol) was dissolved in 88g of toluene, to which 10.0g (0.1mol) of acetylacetone was added slowly with stirring. This produced tri-n-butoxy zirconium acetylacetonate, accompanied by generation of heat. The mixed toluene solution is hereinafter referred to as the PRODUCTION EXAMPLE 3 Catalyst.

PRODUCTION EXAMPLE T4

Tetra-n-butoxy zirconium, 38.3g (0.1mol) was dissolved in 87g of toluene, to which 20.0g (0.2mol) of acetylacetone was added slowly with stirring. This produced di-n-butoxy

zirconiumbisacetylacetonate, accompanied by generation of heat. The mixed toluene solution is hereinafter referred to as the PRODUCTION EXAMPLE 4 Catalyst.

PRODUCTION EXAMPLE T5

5 Tetra-n-butoxy zirconium, 38.8g (0.1mol) was dissolved in 86g of toluene, to which 30.0g (0.3mol) of acetylacetone was added slowly with stirring. This produced di-n-butoxy zirconium trisacetylacetonate, accompanied by generation of heat. The mixed toluene solution is hereinafter referred to
10 as the PRODUCTION EXAMPLE 5 Catalyst.

PRODUCTION EXAMPLE T6

n-Butyl acrylate, 128g (1.0mol), 3.48g (0.015mol) of γ -methacryloxypropylmethyldimethoxysilane, 2.46g (0.015mol) of γ -mercaptopropylmethyldimethoxysilane and 0.25g of
15 α, α' -azobisisobutyronitrile were mixed and dissolved, and 30g of the resultant mixed solution was charged in a 300mL four-mouthed flask purged with nitrogen gas and slowly heated with stirring in an oil bath kept at 70°C. This soon started the polymerization, which was accompanied by generation of heat
20 and thickening of the reaction solution. The remaining mixed solution was slowly added with stirring to the reaction solution dropwise over 2.5 hours via the drip-feed funnel. The reaction solution was continuously stirred for 1 hour after completion of the addition of the mixed solution, to complete the

polymerization. This produced a colorless, transparent, viscous substance having a viscosity of 350P (23°C) at a polymerization rate of 97%.

EXAMPLES T1 to T9

5 The silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), prepared in PRODUCTION EXAMPLE T1, 100 parts by weight, was incorporated with 80 parts by weight of the tackifier resin given in Table T1, to prepare the toluene solution
10 containing the solids at 80% for each of EXAMPLES T1 to T9.

15 The solution was incorporated with the curing catalyst given in Table T1, and the resultant composition was spread over a 25µm thick polyester substrate (Toray Industries, Lumirror Film) by a coater to a thickness of 25µm (on a dry basis), and cured at 120°C for 0.5 to 5 minutes by a drier.

20 The resultant adhesive tape was measured for its releasability from a silicone release paper (Soken Industries, Inc., EK-130R). Its curing speed and resistance to weather were evaluated by the curing speed and weather resistance tests conducted according to the methods described earlier. The results are given in Table T1.

 In Table T1, YS Polyester T-115 and YS Polyester S-145 are terpene phenolic resins (Yasuhara Yushi Kogyo), and Stepelite Ester 7 is a hydrogenated rosin ester resin (Hercules).

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$\text{Zr}(\text{acac})_4$ in Table T1 is zirconium tetraacetylacetonate.

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Table T1

EXAMPLES	Tackifier resin	Curing catalysts		Curing Speed at 120°C	Exfoliation resisting force [g/cm]			Adhesive force (initial) [g/cm]	Residual adhesion rate [%]			Curing speed at room temperature	Resistance to weather *4
		Types	Content (parts by weight)		50°C × 7 days	50°C × 14 days	50°C × 21 days		50°C × 7 days	50°C × 14 days	50°C × 21 days		
T1	YS Polyester T-115	PRODUCTION EXAMPLE 3 Catalyst #1	5	○	4	3	2	430	94	96	92	○	◎
T2	YS Polyester T-115	PRODUCTION EXAMPLE 4 Catalyst #2	5	○	3	5	3	440	92	94	94	○	◎
T3	YS Polyester T-115	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	4	4	3	420	97	99	98	○	◎
T4	YS Polyester S-145	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	4	4	3	800	98	99	96	○	◎
T5	Stepelite Ester 7	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	2	2	3	220	90	88	87	○	◎
T6	YS Polyester T-115	Zr(acac) ₄	5	○	4	3	2	410	86	84	82	○	◎
T7	YS Polyester T-115	(n-BuO) ₄ Zr	5	○	5	5	6	400	87	85	86	○	◎
T8	YS Polyester T-115	Al(acac) ₃	5	○	3	4	4	390	90	88	87	○	◎
T9	YS Polyester T-115	Diisopropoxy Al ethylacetate	5	○	6	6	5	405	88	90	90	○	◎

*1: As (n-BuO)₃Zr(acac), *2: As (n-BuO)₂Zr(acac)₂, *3: As (n-BuO)Zr(acac)₃,

*4: Evaluation standards for resistance to weather, ◎: No change observed, ○: Cracks or molten portion slightly observed, △: Cracks or molten portion observed, and ×: Cracks or molten portion extensively observed

REFERENCE EXAMPLES T1 to T9

The polyalkylene oxide containing the hydrolysable silicon group, prepared in PRODUCTION EXAMPLE T2, 100 parts by weight was incorporated with 80 parts by weight of the tackifier resin given in Table T2, to prepare the toluene solution containing the solids at 80% for each of REFERENCE EXAMPLES T1 to T9.

The solution was incorporated with the curing catalyst given in Table T2, and the resultant composition was spread over a 25 μ m thick polyester substrate (Toray Industries, Lumirror Film) by a coater to a thickness of 25 μ m (on a dry basis), and cured at 120°C for 1 to 19 minutes by a drier.

The resultant adhesive tape was measured for its releasability from a silicone release paper (Soken Industries, Inc., EK-130R). Its curing speed and resistance to weather were evaluated by the curing speed and weather resistance tests conducted according to the methods described earlier. The results are given in Table T2.

In Table T2, YS Polyester T-115 and YS Polyester S-145 are terpene phenolic resins (Yasuhara Yushi Kogyo), and Stepelite Ester 7 is a hydrogenated rosin ester resin (Hercules).

Zr(acac)₄ in Table T2 is zirconium tetraacetylacetonate.

COMPARATIVE EXAMPLES T1 to T3

The adhesive tape was prepared for each of COMPARATIVE EXAMPLES T1 to T3 in the same manner as in COMPARATIVE EXAMPLE T1, except

that the organotin compound as the curing catalyst was used, as shown in Table T2, and measured for its releasability. Its curing speed and resistance to weather were evaluated by the curing speed and weather resistance tests conducted according to the methods described earlier. The results are given in Table T2.

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Table T2

	Tackifier resin	Curing catalysts		Curing Speed at 120°C	Exfoliation resisting force [g/cm]			Adhesive force (initial) [g/cm]	Residual adhesion rate [%]			Curing speed	Resistance to weather #4
		Types	Content (parts by weight)		50°C × 7 days	50°C × 14 days	50°C × 21 days		50°C × 7 days	50°C × 14 days	50°C × 21 days		
REFERENCE EXAMPLE T1	YS Polyster T-115	PRODUCTION EXAMPLE 3 Catalyst #1	5	○	3	3	2	360	90	92	90	○	△
REFERENCE EXAMPLE T2	YS Polyster T-115	PRODUCTION EXAMPLE 4 Catalyst #2	5	○	2	4	3	365	89	92	92	○	△
REFERENCE EXAMPLE T3	YS Polyster T-115	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	2	3	3	360	94	97	95	○	△
REFERENCE EXAMPLE T4	YS Polyster S-145	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	3	3	4	750	92	93	91	○	× to △
REFERENCE EXAMPLE T5	Stepelite Ester 7	PRODUCTION EXAMPLE 5 Catalyst #3	5	○	3	3	3	160	84	80	78	○	△
REFERENCE EXAMPLE T6	YS Polyster T-115	Zr(acac) ₄	5	○	2	3	3	360	74	78	74	○	△
REFERENCE EXAMPLE T7	YS Polyster T-115	(n-BuO) ₄ Zr	5	△	4	4	5	370	77	76	75	○	×
REFERENCE EXAMPLE T8	YS Polyster T-115	Al(acac) ₃	5	○	2	3	3	350	80	78	81	○	△
REFERENCE EXAMPLE T9	YS Polyster T-115	Diisopropoxy Al ethylacetoacetate	5	○	5	6	6	375	78	80	80	○	△
COMPARATIVE EXAMPLE T1	YS Polyster T-115	Dibutyl tin dilaurate	5	×	260	Im [*] measurable	Im [*] measurable	365	50	-	-	×	×
COMPARATIVE EXAMPLE T2	YS Polyster T-115	Dibutyl tin monononyl phenolate	5	○	250	Im [*] measurable	Im [*] measurable	360	34	-	-	○	×
COMPARATIVE EXAMPLE T3	YS Polyster T-115	Dibutyl tin dimethoxide	5	○	240	Im [*] measurable	Im [*] measurable	355	32	-	-	○	×

*1: As (n-BuO)₃Zr(acac), *2: As (n-BuO)₂Zr(acac)₂, *3: As (n-BuO)Zr(acac)₃,

*4: Evaluation standards for resistance to weather, ○: No change observed, △: Cracks or molten portion slightly observed,

△: Cracks or molten portion observed, and ×: Cracks or molten portion extensively observed

As shown in Table T2, the adhesive tape with the composition prepared in each of REFERENCE EXAMPLES T1 to T9 has better releasability from the silicone release paper than that with the composition prepared in each of COMPARATIVE EXAMPLES T1 to

5 T3.

REFERENCE EXAMPLES T10 to T15

The acrylate copolymer containing the hydrolysable silicon group, prepared in PRODUCTION EXAMPLE T6, 100 parts by weight, was incorporated with 50 parts by weight of YS Polyester T-115, to prepare the toluene solution containing the solids at 80% for each of REFERENCE EXAMPLES T10 to T15.

The solution was incorporated with the curing catalyst given in Table T3, and the resultant composition was spread over a 25 μ m thick polyester substrate (Toray Industries, Lumirror Film) to a thickness of 25 μ m (on a dry basis), and cured at 120°C for 3 minutes to prepare the adhesive tape.

The resultant adhesive tape was measured for its releasability from a silicone release paper in the same manner as in COMPARATIVE EXAMPLE T1. Its curing speed and resistance to weather were evaluated by the curing speed and weather resistance tests conducted according to the methods described earlier. The results are given in Table T3.

COMPARATIVE EXAMPLES T4 and T5

The adhesive tape was prepared for each of COMPARATIVE EXAMPLES

T4 and T5 in the same manner as in REFERENCE EXAMPLE T10, except that the curing catalyst was replaced by the organotin compound shown in Table T3, and measured for its releasability. Its curing speed and resistance to weather were evaluated by the curing speed and weather resistance tests conducted according to the methods described earlier. The results are given in Table T3.

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Table T3

	Curing catalysts		Exfoliation resisting force [g/cm]			Adhesive force (initial) [g/cm]	Residual adhesion rate [%]			Curing speed [ordinary temperature]	Resistance to weather #4
	Types	Content (parts by weight)	50°C × 7 days	50°C × 14 days	50°C × 21 days		50°C × 7 days	50°C × 14 days	50°C × 21 days		
REFERENCE EXAMPLE T10	PRODUCTION EXAMPLE 3 Catalyst #1	5	3	3	3	320	84	80	81	○	△ to ×
REFERENCE EXAMPLE T11	PRODUCTION EXAMPLE 4 Catalyst #2	5	3	2	3	330	78	79	81	○	△ to ×
REFERENCE EXAMPLE T12	PRODUCTION EXAMPLE 5 Catalyst #3	5	2	3	3	315	82	82	80	○	△ to ×
REFERENCE EXAMPLE T13	Zr(acac) ₄	5	3	4	4	340	77	76	77	○	△ to ×
REFERENCE EXAMPLE T14	Al(acac) ₃	5	4	6	5	355	79	79	80	○	△
REFERENCE EXAMPLE T15	Diisopropoxy Al ethylacetate	5	3	4	4	345	80	81	81	○	△
COMPARATIVE EXAMPLE T4	Dibutyl tin dilaurate	5	120	250	-	360	45	33	-	○	×
COMPARATIVE EXAMPLE T5	Dibutyl tin monononyphenolate	5	65	170	-	355	38	24	-	○	×

*1: As (n-BuO)₃Zr(acac), *2: As (n-BuO)₂Zr(acac)₂, *3: As (n-BuO)Zr(acac)₃,

*4: Evaluation standards for resistance to weather, ◎: No change observed, ○: Cracks or molten portion slightly observed,

△: Cracks or molten portion observed, and ×: Cracks or molten portion extensively observed

<EXAMPLES U Series>

The composition, the iodine value, the intrinsic viscosity [η] and the molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and REFERENCE EXAPLES were determined by the methods described earlier.

The gel fraction measurement and weather resistance tests in EXAMPLES and REFERENCE EXAMPLES were conducted according to the following methods.

(1) Gel fraction measurement test

The cured coating film was immersed in acetone kept at 20°C for 24 hours, to find the weight of its undissolved portion relative to the film weight before the test. It was marked with × when it was less than 60%, Δ when it was 60% or more but less than 80%, ○ when it was 80% or more but less than 90%, and ◎ when it was 90% or more.

(2) Weather resistance test

The weather resistance test was conducted in accordance with JIS B-7753 using a Sunshine Carbon Arc weatherometer, to determine resistance to weather:

<Testing conditions>

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: 40±2°C

Total light irradiation time: 250 hours

<Evaluation standards for resistance to weather>

○ : No cracks or molten portion observed on one side of the tested piece

5 △ : Cracks or molten portion observed slightly on one side of the tested piece

× : Cracks or molten portion observed on one side of the tested piece

EXAMPLE U

10 [Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

15 The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$ 20 and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an iodine value of 10(g/100g), an intrinsic viscosity $[\eta]$, as measured in decalin at 135°C, of 0.2dl/g, and Mw/Mn of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-SiCH₃(OCH₃)₂).

REFERENCE EXAMPLE U1

A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 16g of allylmethacrylate, 20g of methylmethacrylate, 19g of n-butyl methacrylate, 14g of butyl acrylate, 4g of maleic

anhydride and 2g of n-dodecylmercaptan was added dropwise in 90g of xylene as the solvent kept at 90°C, and these compounds were allowed to react with each other for 10 hours, to obtain the vinyl-based polymer having a molecular weight of 8,000 and containing an allyl type unsaturated group. The polymer had absorptions relevant to the carbon-carbon double bond and acid anhydride at 1648 and 1780cm⁻¹, respectively, in the infrared absorption spectral pattern. The polymer solution was distilled under a vacuum to remove 40g of the solvent.

A solution of 1.5g of trimethoxysilane and 0.0005g of chloroplatinic acid dissolved in isopropanol was added to 16g of the vinyl-based polymer having an allyl type unsaturated group, and they were allowed to react with each other at 90°C for 6 hours in a sealed system. The reaction product had no infrared absorption at 1648cm⁻¹ in the infrared absorption spectral pattern, from which it was judged that silyl-containing vinyl-based polymer was produced.

REFERENCE EXAMPLE U2

A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 22g of γ-methacryloxypropyltrimethoxysilane, 22g of methyl methacrylate, 15g of n-butyl methacrylate and 18g of butyl acrylate was added dropwise in 70g of xylene as the solvent kept at 90°C, and these compounds were allowed to react with each other for 10 hours, to obtain the silyl-containing

vinyl-based polymer having a molecular weight of 16,000.

REFERENCE EXAMPLE U3

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5 A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 22g of γ -methacryloxypropyltrimethoxysilane, 22g of methyl methacrylate, 15g of n-butyl methacrylate, 18g of butyl acrylate and 2g of n-dodecylmercaptan was added dropwise in 70g of xylene as the solvent kept at 90°C, and these compounds were allowed to react with each other for 10 hours, to obtain the silyl-containing vinyl-based polymer having a molecular weight
10 of 9,000.

REFERENCE EXAMPLE U4

15 A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 22g of γ -methacryloxypropyltrimethoxysilane, 52g of methyl methacrylate, 15g of n-butyl methacrylate, 18g of butyl acrylate, 4g of acrylamide, 10g of n-butanol and 4g of n-dodecylmercaptan was added dropwise in 70g of xylene as the solvent kept at 70°C, and these compounds were allowed to react with each other for 10 hours, to obtain the silyl-containing vinyl-based polymer having a molecular weight of 6,000.

20 REFERENCE EXAMPLE U5

A solution of 2g of azobisisobutyronitrile dissolved in 30g of styrene, 22g of γ -methacryloxypropyltrimethoxysilane, 22g of methyl methacrylate, 15g of n-butyl methacrylate, 18g of butyl acrylate, 4g of 2-hydroxyethyl methacrylate and 4g of

n-dodecylmercaptan was added dropwise in 70g of xylene as the solvent kept at 90°C, and these compounds were allowed to react with each other for 10 hours, to obtain the silyl-containing vinyl-based polymer having a molecular weight of 6,000.

5 REFERENCE EXAMPLE U6

10 The silyl-containing vinyl-based polymer having a molecular weight of 5,000 was obtained in the same manner as in REFERENCE EXAMPLE U4, except that 4g of n-dodecylmercaptan and 4g of 2-hydroxyethyl methacrylate in REFERENCE EXAMPLE U5 were replaced by 6g of n-dodecylmercaptan, 4g of acrylamide, 2g of maleic anhydride and 10g of n-butanol.

15 The resin solution obtained in each of EXAMPLE U and REFERENCE EXAMPLES U1 to U6 was incorporated with the additive(s) and the curing catalyst(s) given in Table U1, and diluted with xylene to the resin viscosity (Ford cup viscosity: 15 seconds), to measure the pot life before it was skinned or gelled under open conditions.

20 Moreover, the gel fraction measurement test was conducted in accordance with the above-described method for the mixture of the resin solution, obtained in each of EXAMPLE U and REFERENCE EXAMPLES U1 to U6, and the additive(s) and the curing catalyst(s) given in Table U1.

The mixture was left in a glass petri dish, 2cm in diameter and 1.5cm deep, to be cured at room temperature, and the cured

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composition was measured for resistance to weather by the above-described method.

The results are given in Table U1.

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Table U1

	EXAMPLE U	REFERENCE EXAMPLE U1	REFERENCE EXAMPLE U2	REFERENCE EXAMPLE U3		REFERENCE EXAMPLE U4	REFERENCE EXAMPLE U5		REFERENCE EXAMPLE U6
				(1)	(2)		(1)	(2)	
Curing catalyst [parts by weight]	3	3	3	2.4	-	3	-	-	3
Stann JF-9B (*1)	-	-	-	-	-	-	-	1	-
Phthalic acid	-	-	-	-	-	-	3	3	-
Dibutyl tin dilaurate	-	-	-	-	-	-	-	-	-
Tin octylate	-	-	-	0.6	1	-	-	-	-
Additive [parts by weight]									
Methanol	10	10	10	-	-	-	10	10	10
Tetraethyl orthosilicate	10	10	-	10	10	-	10	10	10
Trimethyl orthoformate	-	-	-	-	-	-	-	-	1
Pot life under open conditions	10hrs \leq	10hrs \leq	10hrs \leq	10hrs \leq	Skimming in 3hrs	10hrs \leq	Skimming in 3hrs	10hrs \leq	10hrs \leq
Evaluation of gel fraction	⊙	×	△	×	○	△	○	×	△
Evaluation of resistance to weather	○	△	△	△	×	△	×	△	×

(*1) Stann JF-98: Stabilizer for vinyl chloride (Sankyo Organic Chemicals, Ltd.)

Chemical formula of the major ingredient: $(n\text{-C}_4\text{H}_9)_2\text{Sn}(\text{-SCH}_2\text{COOR})_2$, (R: C4 to C12)

(*2) Parts by weight for the curing catalyst and additive are based on 100 parts by weight of the resin

<EXAMPLES V Series>

The composition, the iodine value, the intrinsic viscosity $[\eta]$ and the molecular weight distribution (M_w/M_n) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAPLES were determined by the methods described earlier.

PRODUCTION EXAMPLE V1

[Production of silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $VO(OEt)_2Cl$ and $Al(Et)_{1.5}Cl_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent

by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1) thus produced contained ethylene at 68% by mol, and had an iodine value of 10(g/100g), an intrinsic viscosity $[\eta]$, as measured in decalin at 135°C, of 0.2dl/g, and Mw/Mn of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-SiCH₃(OCH₃)₂).

PRODUCTION EXAMPLE V2

[Production of saturated hydrocarbon-based polymer (D-1)]

A uniformly mixed solution of 560mL of methylene chloride, 1,160mL of n-hexane, 940mg of α -methylpyridine and 22g of p-dicumyl chloride, all dried, was formed in a four-mouthed flask equipped with an agitator and nitrogen line and cooled to -70°C, in which 570mL of isobutylene monomer was charged under a vacuum

through a molecular sieves tube.

A polymerization catalyst solution (comprising 14mL of titanium tetrachloride and 80mL of methylene chloride) cooled beforehand was added all at once to the above reaction solution, kept at -70°C , with stirring to initiate the polymerization reaction. The reaction solution was heated to -54°C , and then cooled to -70°C in about 17 minutes.

About 20 minutes after the polymerization was initiated, 132g of 1,9-decadiene was added to the reaction solution, and continuously stirred at -70°C for 4 hours.

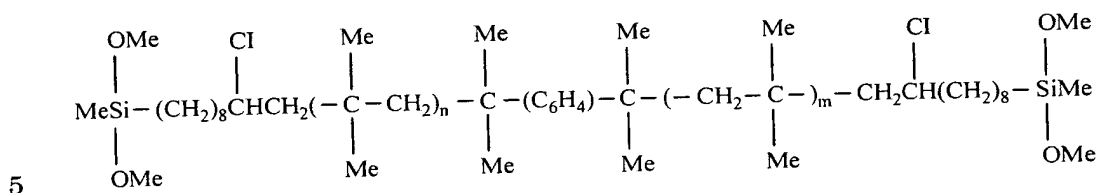
The yellowy turbid reaction solution thus produced was put in 3L of warm water (around 45°C) and stirred for around 2 hours. Then, the organic layer was separated, and washed with pure water 3 times. The resultant colorless, transparent organic layer was concentrated under a vacuum, to obtain approximately 400g of the isobutylene oligomer with vinyl groups at both terminals.

Next, 400g of the isobutylene oligomer with vinyl groups at both terminals was dissolved in 200mL of n-heptane. The resultant solution was heated to around 70°C , to which 1.5 [eq/vinyl group] of methyl dimethoxysilane and 1×10^{-4} [eq/vinyl group] of a platinum/vinyl siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 4 hours.

The reaction solution was concentrated under a vacuum, to

obtain the target isobutylene oligomer (D-1) having the reactive silicon group at both terminals.

[Structural formula]



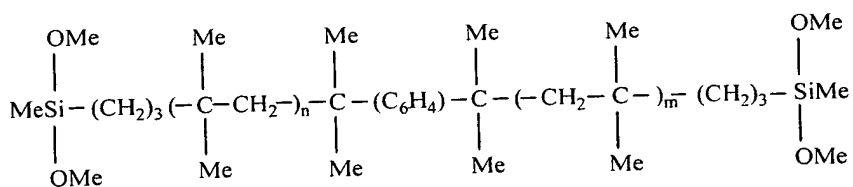
PRODUCTION EXAMPLE V3

[Production of saturated hydrocarbon-based polymer (D-2)]

10 The same procedure as used for PRODUCTION EXAMPLE V2 was repeated in PRODUCTION EXAMPLE V3, except that 1,9-decadiene was replaced by 24g of allylmethylsilane, to obtain the isobutylene oligomer (D-2) of partly different production intermediate structure.

[Structural formula]

15



PRODUCTION EXAMPLE V4

[Production of saturated hydrocarbon-based polymer (D-3)]

20 A uniformly mixed solution of 560mL of methylene chloride,

1,160mL of n-hexane, 940mg of α -methylpyridine and 22g of p-dicumyl chloride, all dried, was formed in a four-mouthed flask equipped with an agitator and nitrogen line, and cooled to -70°C , in which 570mL of isobutylene monomer was charged under a vacuum
5 through a molecular sieves tube.

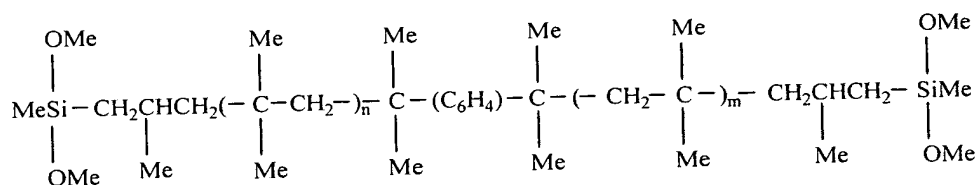
A polymerization catalyst solution (comprising 14mL of titanium tetrachloride and 80mL of methylene chloride) cooled beforehand was added all at once to the above reaction solution, kept at -70°C , with stirring to initiate the polymerization
10 reaction. The reaction solution was heated to -54°C , and then cooled to -70°C in about 17 minutes. The reaction solution was continuously stirred for around 60 minutes after the polymerization was initiated. The yellowy turbid reaction
15 solution thus produced was put in 3L of warm water (around 45°C) and stirred for around 2 hours. Then, the organic layer was separated, and washed with pure water 3 times. The resultant colorless, transparent organic layer was concentrated under a vacuum, to obtain approximately 400g of the isobutylene oligomer with chlorous groups at both terminals.

20 Then, the isobutylene oligomer was continuously heated at 170°C under a vacuum for 2 hours for the thermal dehydrochlorination reaction, to obtain the isobutylene oligomer with isopropenyl groups at both terminals.

Next, 400g of the isobutylene oligomer with isopropenyl groups

at both terminals, prepared above, was dissolved in 200mL of n-heptane. The resultant solution was heated in a pressure vessel to around 100°C, to which 1.5 [eq/vinyl group] of methyl dichlorosilane and 1×10^{-4} [eq/vinyl group] of a platinum/vinyl siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 10 hours. The reaction solution was cooled to 60°C, to which an excess quantity of methanol over methyl dichlorosilane was added, and the mixture was stirred for around 4 hours, to complete the methoxylation. The reaction solution was concentrated under a vacuum, to obtain the target isobutylene oligomer (D-3) having the structure with the reactive silicon group at both terminals.

[Structural formula]



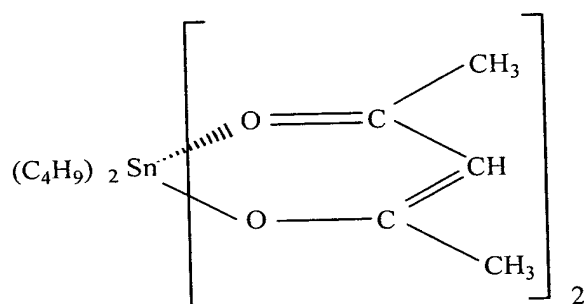
EXAMPLE V1

A mixture containing the polymer (A-1) prepared in PRODUCTION EXAMPLE V1 as the silane-modified ethylene/ α -olefin/non-conjugated polyene random copolymer rubber (A1) was prepared. It was composed of 100 parts of the

polymer rubber (A-1), 120 parts of calcium carbonate (Shiraishi K.K., CCRTM), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM) as the plasticizer, 30 parts of titanium dioxide, 5 parts of sodium bisulfate, and 3 parts of dibutyl tin bisacetylacetonate as the curing catalyst (H8), all parts by weight. These components were uniformly kneaded to obtain the curable composition.

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was cured in 16 minutes.

Dibutyl tin bisacetylacetonate



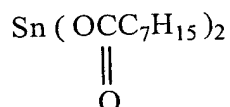
15 COMPARATIVE EXAMPLE V1

The curable composition was prepared in the same manner as in EXAMPLE V1, except that the curing catalyst (H8) was replaced by tin dioctylate.

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was not cured even

after a lapse of 700 minutes or more.

Tin dioctylate



5

REFERENCE EXAMPLE V1

10 A mixture containing the polymer (D-1) prepared in PRODUCTION
EXAMPLE V2 as the saturated hydrocarbon-based polymer component
was prepared. It was composed of 100 parts of the polymer (D-1),
120 parts of calcium carbonate (Shiraishi K.K., CCRTM), 90 parts
of a paraffin-based process oil (Idemitsu Kosan, Diana Process
Oil PS-32TM) as the plasticizer, 30 parts of titanium dioxide,
5 parts of sodium bisulfate, and 3 parts of dibutyl tin
15 bisacetylacetonate as the curing catalyst (H8), all parts by
weight. These components were uniformly kneaded to obtain the
curable composition.

Curability (tack-free time) of the resultant composition was
measured in accordance with JIS A-5758. It was cured in 25
minutes.

20 REFERENCE EXAMPLE V2

A mixture containing the polymer (D-2) prepared in PRODUCTION
EXAMPLE V3 as the saturated hydrocarbon-based polymer component
was prepared. It was composed of 100 parts of the polymer (D-2),

120 parts of calcium carbonate (Shiraishi K.K., CCRTM), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM) as the plasticizer, 30 parts of titanium dioxide, 5 parts of sodium bisulfate, and 3 parts of dibutyl tin bisacetylacetonate as the curing catalyst (H8), all parts by weight. These components were uniformly kneaded to obtain the curable composition.

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was cured in 30 minutes.

REFERENCE EXAMPLE V3

A mixture containing the polymer (D-3) prepared in PRODUCTION EXAMPLE V4 as the saturated hydrocarbon-based polymer component was prepared. It was composed of 100 parts of the polymer (D-3), 120 parts of calcium carbonate (Shiraishi K.K., CCRTM), 90 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM) as the plasticizer, 30 parts of titanium dioxide, 5 parts of sodium bisulfate, and 3 parts of dibutyl tin bisacetylacetonate as the curing catalyst (H8), all parts by weight. These components were uniformly kneaded to obtain the curable composition.

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was cured in 30 minutes.

REFERENCE EXAMPLE V4

The curable composition was prepared in the same manner as in REFERENCE EXAMPLE V1, except that the polymer (D-1) prepared in PRODUCTION EXAMPLE V2 as the saturated hydrocarbon-based polymer was incorporated with tin dioctylate in place of the curing catalyst (H8).

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was not cured even after a lapse of 700 minutes or more.

REFERENCE EXAMPLE V5

The curable composition was prepared in the same manner as in REFERENCE EXAMPLE V2, except that the polymer (D-2) prepared in PRODUCTION EXAMPLE V3 as the saturated hydrocarbon-based polymer was incorporated with tin dioctylate in place of the curing catalyst (H8).

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was not cured even after a lapse of 700 minutes or more.

REFERENCE EXAMPLE V6

The curable composition was prepared in the same manner as in REFERENCE EXAMPLE V3, except that the polymer (D-3) prepared in PRODUCTION EXAMPLE V4 as the saturated hydrocarbon-based polymer was incorporated with tin dioctylate in place of the curing catalyst (H8).

Curability (tack-free time) of the resultant composition was measured in accordance with JIS A-5758. It was not cured even after a lapse of 700 minutes or more.

The curing speed test and accelerated weather resistance test
5 were conducted by the following methods for EXAMPLES and
COMPARATIVE EXAMPLES.

(1) Curing speed test

The curable composition was cured under the conditions of
23°C and 50%RH for 24 hours in a mold, 20 by 80 by 5mm in size.

10 Next, the cured product was released from the mold, and
thickness of the cured portion was measured by a dial gauge of
weak spring force to 0.1mm, to evaluate its curing speed. It
was marked with ○ when its thickness was 1mm or more, and ×
when it was less than 1mm.

15 (2) Weather resistance test

The accelerated weather resistance test was conducted in
accordance with JIS B-7753 under the following conditions:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120

20 minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 250 hours

The tested test piece was visually observed, and marked with

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○ when no deterioration (cracks or molten portion) was observed,
and × when the deterioration was observed.

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Table V1

	EXAMPLE	COMPARATIVE EXAMPLE
	V1	V1
Silyl-containing ethylene/ α -olefin/non-conjugated polyethylene copolymer rubber (A-1)	100	100
	3	-
Curing catalyst	-	3
	Curing catalyst (H8) Tin dioctylate	
Calcium carbonate	120	120
Process oil	90	90
Titanium dioxide	30	30
Sodium hydrogen sulfate	5	5
Tack-free (min)	16	>700
Curability	○	×
Resistance to weather	○	○

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Table V2

		REFERENCE EXAMPLE V					
		1	2	3	4	5	6
Saturated hydrocarbon-based Polymer	D-1	100	-	-	100	-	-
	D-2	-	100	-	-	100	-
	D-3	-	-	100	-	-	100
	Curing catalyst (H8)	3	3	3	-	-	-
Curing catalyst	Tin dioctylate	-	-	-	3	3	3
		120	120	120	120	120	120
Calcium carbonate		90	90	90	90	90	90
Process oil		30	30	30	30	30	30
Titanium dioxide		5	5	5	5	5	5
Sodium hydrogen sulfate		25	30	30	>700	>700	>700
Tack-free (min)		×	×	×	×	×	×
Curability		○	○	○	○	○	○
Resistance to weather							

<EXAMPLES W Series>

The composition, the iodine value, the intrinsic viscosity $[\eta]$ and the molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE
 5 EXAMPLES were determined by the methods described earlier.

Curing speed and weather resistance of the compositions prepared in EXAMPLES, and COMPARATIVE and REFERENCE EXAMPLES were determined by the following methods.

(1) Curing speed test

10 The curable composition was cured at 23°C and 50%RH in a mold (80 by 80 by 12.5mm in size), wherein hardness was followed by a JIS-A hardness meter after surface tackiness disappeared, to record the time required for hardness to reach 20.

(2) Weather resistance test

15 Accelerated weather resistance test:

Conducted in accordance with JIS B-7753.

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

20 Black panel temperature: $63 \pm 2^\circ\text{C}$

Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 500 hours

PRODUCTION EXAMPLE W1

[Production of silyl-containing

ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having
5 an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OEt})_2\text{Cl}$
10 and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0 -1) in a form of uniform solution.

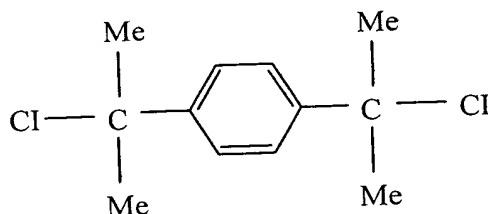
15 A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

20 The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0 -1) thus produced contained ethylene at 68% by mol, and had an iodine value of 10 (g/100g), intrinsic viscosity $[\eta]$, as measured in decalin kept at 135°C, of 0.2dl/g, and M_w/M_n of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) containing dimethoxymethylsilyl group (-Si(CH₃)(OCH₃)₂).

PRODUCTION EXAMPLE W2

A 2L pressure-resistant glass reactor was charged, after it was equipped with a 3-way cock and purged with nitrogen, with 138mL of ethyl cyclohexane (dried by molecular sieves 3A at least for a night), 1012mL of toluene (also dried by molecular sieves 3A at least for a night) and 8.14g (35.2mmols) of p-DCC represented by the following formula by a syringe.



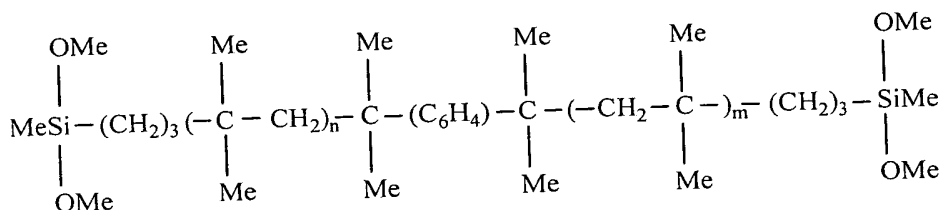
Next, a pressure-resistant glass-made liquefied gas

collecting tube equipped with a needle valve and containing 254mL (2.99mols) of an isobutylene monomer was connected to one side of the 3-way cock. Then the reactor for polymerization was immersed in a dry ice/ethanol bath kept at -70°C to cool the solution, and evacuated to a vacuum using a vacuum pump. It was then charged with the isobutylene monomer from the liquefied gas collecting tube by opening the needle valve, and returned back to the normal pressure with a nitrogen gas introduced from another side of the 3-way cock. The reactor was charged with 0.387g (4.15mmols) of 2-methyl pyridine and then with 4.90mL (44.7mmols) of titanium tetrachloride, to initiate the polymerization. After a lapse of 70 minutes, 9.65g (13.4mmols) of allyl trimethylsilane was added to the reactor to introduce the allyl group at the polymer terminal. After a lapse of 120 minutes for the reaction, the reaction solution was washed 4 times each with 200mL of water, and the solvent was distilled off to obtain the isobutylene-based polymer with the allyl group at the terminal.

Next, 200g of the isobutylene-based polymer with the allyl group at the terminal thus obtained was mixed with 60g of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM) as the hydrocarbon-based plasticizer, and the mixture was heated to around 75°C , to which 1.5 [eq/vinyl group] of methyl dimethoxysilane and 5×10^{-5} [eq/vinyl group] of a platinum/vinyl

siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 20 hours.

This produced the mixture of the isobutylene polymer (represented by the following formula) having the reactive silicon group at both terminals and PS-32 as the plasticizer (10/3 by weight).



10

The polymer yield was estimated from the quantity produced. It was also analyzed for Mn and Mw/Mn by GPC, and the terminal structure by comparing the intensities of the 300MHz ^1H -NMR-analyzed resonance signals of proton relevant to each structure (proton derived from the initiator: 6.5 to 7.5ppm, methyl proton bonded to the silicon atom, derived from the polymer terminal: 0.0 to 0.1ppm, and methoxy proton: 3.4 to 3.5ppm) with each other. The ^1H -NMR analysis was conducted using a Varian Gemini 300 (300MHz for ^1H) in CDCl_3 .

20

The FT-IR analysis was conducted by an IR analyzer (Shimadzu IR-408), and GPC analysis was conducted with a Waters LC Module 1 as the liquid sending system and Shodex K-804 as the column.

The molecular weight was the one relative to the polystyrene standard. The polymer thus prepared had an Mn of 5,780, Mw/Mn of 1.28 and Fn (silyl) of 1.93, wherein the number-average molecular weight was as polystyrene, and the number of the
5 terminal functional silyl group was that per 1 mol of isobutylene polymer.

EXAMPLE W1

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The silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1), 130 parts, produced in PRODUCTION EXAMPLE W1, and a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32™), in which the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber as the component (A-1) accounted for 100 parts, was
15 incorporated with 5 parts of tetra-n-butyl titanate (Wako-Junyaku) as the titanates of the component (Y), 2 parts of dibutyl tin bisacetylacetonate (NITTO KASEI, Neostann U-220™) as the silanol condensing catalyst and 1 part of H₂O, all parts by weight, to prepare the curable rubber composition.

20 The above composition was spread over a float glass substrate washed with methylethylketone (Wako-Junyaku), which was not coated with a primer, to a thickness of 5mm, and cured in an oven. The cured composition was peeling-tested, in which it was manually released while the adhesive surface being cut by

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a cutter knife. The composition containing 5 parts by weight of the titanate was well adhesive to the float glass substrate, showing the cohesion fracture. The curing speed and weather resistance tests were also conducted according to the methods described earlier. It took 18 hours until its hardness reached 20 in the curing speed test, and showed no cracks in the weather resistance test.

COMPARATIVE EXAMPLE W1

The composition was prepared in the same manner as in EXAMPLE W1, except that addition of the titanates as the component (Y) was omitted, and tested in the same manner. It was insufficient in adhesion to the float glass substrate, showing the adhesion fracture. Moreover, it took 72 hours or more until its hardness reached 20 in the curing speed test, and showed a number of cracks visible to the naked eye in the weather resistance test.

REFERENCE EXAMPLE W1

The composition was prepared in the same manner as in COMPARATIVE EXAMPLE W1, except that the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber prepared in PRODUCTION EXAMPLE W1 was replaced by the saturated hydrocarbon-based polymer containing the reactive silicon group prepared in PRODUCTION EXAMPLE W2, and tested in the same manner. The manual peeling test results indicated that the composition was insufficient in adhesion to the float glass

substrate, showing the adhesion fracture. Moreover, it took 48 hours until its hardness reached 20 in the curing speed test, and showed cracks and molten portion visible to the naked eye, although slightly, in the weather resistance test.

5 REFERENCE EXAMPLE W2

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The composition was prepared in the same manner as in EXAMPLE W1, except that the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber prepared in PRODUCTION EXAMPLE W1 was replaced by the saturated hydrocarbon-based polymer containing the reactive silicon group prepared in PRODUCTION EXAMPLE W2, and tested in the same manner. The manual peeling test results indicated that the composition was good in adhesion to the float glass substrate, showing the cohesion fracture. However, it took 36 hours until its hardness reached 20 in the curing speed test, and showed cracks and molten portion, although slightly, in the weather resistance test.

15 <EXAMPLES X Series>

The composition, the iodine value, the intrinsic viscosity [η] and the molecular weight distribution (Mw/Mn) of the copolymer rubber used in each of EXAMPLES and COMPARATIVE EXAMPLES were determined by the methods described earlier.

20 PRODUCTION EXAMPLE X1

[Production of silyl-containing

ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1)]

The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 3.0kg, 9.0kg and 550g per hour, respectively, from the reactor side into the liquid phase, and hydrogen 70L, VOCl_3 , $\text{Al}(\text{Et})_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 95mmols, 443 mmols and 127mmols per hour, respectively, also continuously.

The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0 -1) in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A_0 -1) thus produced contained ethylene at 68% bymol, and had an iodine value of 15(g/100g), intrinsic viscosity $[\eta]$, as measured in decalin at 135°C, of 0.2dl/g, and M_w/M_n of 15. The yield was 3.5kg/h.

Two% toluene solution (0.3 part by weight) of chloroplatinic acid and 1.5 parts by weight of methyldimethoxysilane were added to 100 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A₀-1), and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent (toluene) were distilled off from the effluent. This produced 101.5 parts by weight of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber containing dimethoxymethylsilyl group (-SiH(OCH₃)₂).

PRODUCTION EXAMPLE X2

[Production of saturated hydrocarbon-based polymer (CA-1)]

A uniformly mixed solution of 560mL of methylene chloride, 1,160mL of n-hexane, 940mg of α-methylpyridine and 22g of p-dicumyl chloride, all dried, was formed in a four-mouthed flask equipped with an agitator and nitrogen line and cooled to -70°C, in which 570mL of isobutylene monomer was charged under a vacuum through a molecular sieves tube.

A polymerization catalyst solution (comprising 14mL of titanium tetrachloride and 80mL of methylene chloride) cooled beforehand was added all at once to the above reaction solution, kept at -70°C, with stirring to initiate the polymerization reaction. The reaction solution was heated to -54°C, and then cooled to -70°C in about 17 minutes. About 20 minutes after

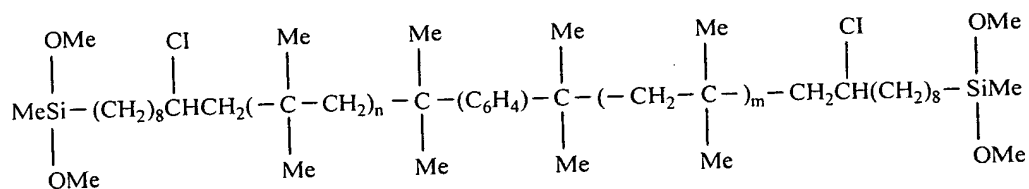
the polymerization was initiated, 132g of 1,9-decadiene was added to the reaction solution, and continuously stirred at -70°C for 4 hours.

The yellowy turbid reaction solution thus produced was put in 3L of warm water (around 45°C) and stirred for around 2 hours. Then, the organic layer was separated, and washed with pure water 3 times.

The resultant colorless, transparent organic layer was concentrated under a vacuum, to obtain approximately 400g of the isobutylene oligomer with vinyl groups at both terminals.

Next, 400g of the isobutylene oligomer with vinyl groups at both terminals was dissolved in 200mL of n-heptane. The resultant solution was heated to around 70°C , to which 1.5 [eq/vinyl group] of methyl dimethoxysilane and 1×10^{-4} [eq/vinyl group] of a platinum/vinyl siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 4 hours.

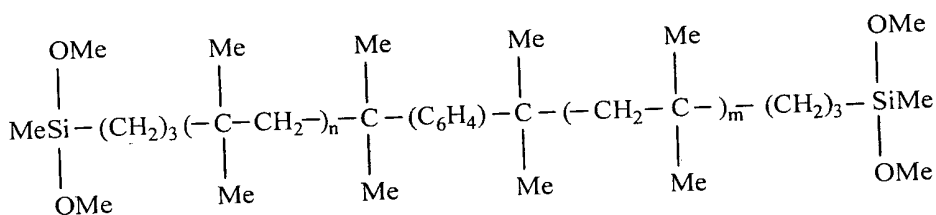
The reaction solution was concentrated under a vacuum, to obtain the isobutylene oligomer (represented by the following formula) having the reactive silicon group at both terminals.



PRODUCTION EXAMPLE X3

[Production of saturated hydrocarbon-based polymer (CA-2)]

The same procedure as used for PRODUCTION EXAMPLE X2 was
 5 repeated in PRODUCTION EXAMPLE X3, except that 1,9-decadiene
 was replaced by 24g of allylmethylsilane, to obtain the
 isobutylene oligomer (represented by the following formula) of
 a partly different production intermediate structure.

PRODUCTION EXAMPLE X4

[Production of saturated hydrocarbon-based polymer (CA-3)]

A uniformly mixed solution of 560mL of methylene chloride,
 15 1,160mL of n-hexane, 940mg of α -methylpyridine and 22g of
 p-dicumyl chloride, all dried, was formed in a four-mouthed flask
 equipped with an agitator and nitrogen line and cooled to -70°C ,
 in which 570mL of isobutylene monomer was charged under a vacuum
 through a molecular sieves tube.

20 A polymerization catalyst solution (comprising 14mL of
 titanium tetrachloride and 80mL of methylene chloride) cooled
 beforehand was added all at once to the above reaction solution,

kept at -70°C , with stirring to initiate the polymerization reaction. The reaction solution was heated to -54°C , and then cooled to -70°C in about 17 minutes. The reaction solution was continuously stirred for around 60 minutes after the

5 polymerization was initiated. The yellowy turbid reaction solution thus produced was put in 3L of warm water (around 45°C) and stirred for around 2 hours. Then, the organic layer was separated, and washed with pure water 3 times. The resultant colorless, transparent organic layer was concentrated under a

10 vacuum, to obtain approximately 400g of the isobutylene oligomer with chlorous groups at both terminals.

Then, the isobutylene oligomer was continuously heated at 170°C under a vacuum for 2 hours for the thermal dehydrochlorination reaction, to obtain the isobutylene

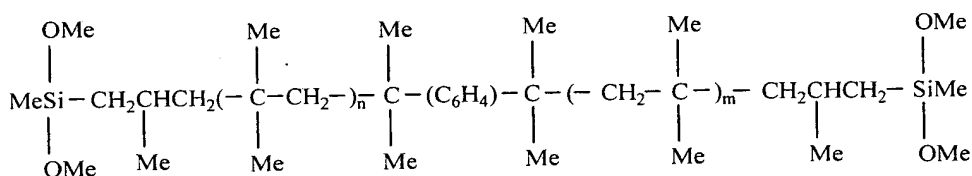
15 oligomer with isopropenyl groups at both terminals.

Next, 400g of the isobutylene oligomer with isopropenyl groups at both terminals was dissolved in 200mL of n-heptane. The resultant solution was heated in a pressure vessel to around 100°C , to which 1.5 [eq/vinyl group] of methyl dichlorosilane

20 and 1×10^{-4} [eq/vinyl group] of a platinum/vinyl siloxane complex were added, for the hydrosilylation. The reaction was followed by FT-IR. The olefin absorption at 1640cm^{-1} disappeared in around 10 hours. The reaction solution was cooled to 60° , to which an excess quantity of methanol over methyl dichlorosilane was

added, and the mixture was stirred for around 4 hours, to complete the methoxylation. The reaction solution was concentrated under a vacuum, to obtain the isobutylene oligomer having the structure represented by the following formula, with the reactive

5 silicon group at both terminals.



SYNTHESIS EXAMPLE 1

10 [Synthesis of copolymer (B)]

A solution dissolving 5.7g of butyl acrylate, 65.1g of methyl methacrylate, 13.3g of stearyl methacrylate, 5.6g of γ -methacryloxypropyltrimethoxysilane, 8.0g of γ -mercaptopropyltrimethoxysilane, 5.0g of

15 azobisisobutyronitrile and 22g of xylene was added dropwise to 20g of xylene as the solvent heated at 110°C over 6 hours. They were allowed to react for polymerization with each other for 2 hours, to obtain the copolymer (B) containing the solids at 70% and having a number-average molecular weight (Mn) of 2,100

20 as polystyrene, determined by GPC.

EXAMPLES X1 to X5

The silyl-containing

ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) prepared in PRODUCTION EXAMPLE X1 was blended with the copolymer (B) prepared in SYNTHESIS EXAMPLE 1 in a solid ratio of 60/40 by weight, and the mixture was evaporation-treated at 110°C under a vacuum by an evaporator, to obtain the transparent, viscous liquid containing the solids at 99% or more.

The blended, evaporation-treated polymer, 100g, was thoroughly kneaded with 100g of limestone powder, 50g of colloidal calcium carbonate, 5g of glass balloons (average particle size: 70 μ m), 100g of diisononyl phthalate, 5g of silicic anhydride, 2g of a hindered phenol-based aging inhibitor, 10g of calcium oxide, 2g of an aluminum chelate-based curing catalyst, 1g of an aminosilane compound and a silicone-based reactive diluent (Shin-etsu Silicone, AFP-1) by a planetary mixer, to obtain the sample (A-1) for EXAMPLES X1 to X5.

REFERENCE EXAMPLES X1 to X9

The blended, evaporation-treated polymers were prepared for REFERENCE EXAMPLES X1 to X9 in the same manner as in EXAMPLES X1 to X5, except that the silyl-containing ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber (A-1) prepared in PRODUCTION EXAMPLE X1 was replaced by the saturated hydrocarbon-based polymers (CA-1) to (CA-3) prepared in PRODUCTION EXAMPLES X2 to X4, respectively, and the samples (CA-1) to (CA-3) were prepared for COMPARATIVE EXAMPLES

X1 to X3 in the same manner as in EXAMPLES X1 to X5.

COMPARATIVE EXAMPLES X1 to X3

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A vinyl chloride resin, 100g, was thoroughly kneaded with 100g of limestone powder, 50g of colloidal calcium carbonate, 5g of glass balloons (average particle size: 70 μ m), 100g of diisononyl phthalate, 3g of lead-based dehydrochlorination inhibitor and 5g of urethane prepolymer by a planetary mixer, to obtain the sample (CA-4) for COMPARATIVE EXAMPLES X1 to X3.

Each of these samples was spread over a cation-electrodeposited steel plate to a thickness given in Table X1 under varying conditions also given in Table X1, and cured to investigate its resistance to chipping, sprayed saline water and vibration. Resistance to weather and curing speed of the coating film were evaluated by the following methods. The results are given in Table X1.

The test methods are described below.

(Resistance to chipping)

Three types of nuts (Nut M-4) were dropped onto the sample slanted at 45° from a height of 2m until the base was exposed, and the chipping resistance was evaluated by the total weight of the nuts.

(Resistance to sprayed saline water)

The sample coated on the cation-electrodeposited steel plate as the film of a given thickness was cross-cut at the center

to the base metal, and put in a tank sprayed with saline water for 200 hours. The resistance to sprayed saline water was evaluated by the maximum width of exfoliation, when a cellophane tape put on the cut surface was torn off.

5 (Resistance to vibration)

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The resistance to vibration was evaluated by the vibration insulation coefficient (d), defined by the formula $d=(f_2-f_1)/f_0$ in accordance with the vibration insulation test method for automobile underbody coating materials (JASO7006), wherein f_0 is the resonance frequency at the secondary resonance point, and f_2 and f_1 are frequencies at the sections where sound intensity is decreased by 3dB, the frequencies being measured at 25°C.

(Curing speed test)

Tackiness-free time at medium temperature

15 A cation-electrodeposited steel plate coated with the composition to a thickness of 10mm was kept at 60°C in a drier. Tackiness of the coating composition was measured at given time intervals by fingering. Tackiness-free time is defined as the time at which the composition is no longer transferred to the
20 fingertip.

(Weather resistance test)

The weather resistance was evaluated by the accelerated weather resistance test conducted in accordance with JIS B-7753 under the following conditions:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120
minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^\circ\text{C}$

5 Tank inside temperature: $40 \pm 2^\circ\text{C}$

Total light irradiation time: 250 hours

The tested test piece was visually observed, to evaluate its
resistance to weather according to the following four grades:

◎: No cracks or molten portion observed entirely

10 ○: Cracks or molten portion observed very slightly

△: Cracks or molten portion observed to some extent

×: Cracks or molten portion observed massively

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Table XI

EXAMPLE X										REFERENCE EXAMPLE X										COMPARATIVE EXAMPLE X		
A-1										CA-1			CA-2			CA-3				CA-4		
1	2	3	4	5	1	2	3	4	5	6	7	8	9	1	2	3						
Curing condition	140°C×30min	○	○	-	-	-	○	○	-	○	○	○	-	○	○	-						
	120°C×20min	-	-	○	-	-	○	-	○	-	○	-	○	-	-	○						
	120°C×10min	-	-	-	○	-	-	-	-	-	-	-	-	-	-	-						
	100°C×20min	-	-	-	-	○	-	-	-	-	-	-	-	-	-	-						
Thickness of coating film	1.5mm	○	-	-	-	-	-	○	-	-	○	-	-	○	-	-						
	0.5mm	-	○	○	○	○	-	-	○	○	-	○	○	-	○	○						
Resistance to chipping		135	98	92	92	81	103	79	74	102	80	79	101	73	77	68	41	9				
Resistance to vibration (×10³)		55	16	15	14	14	45	10	12	42	11	12	42	11	13	8	5	4				
Resistance to sprayed saline water (mm)		≤1	≤1	≤1	≤1	≤1	≤1	≤1	1.2	≤1	≤1	1.3	≤1	≤1	1.3	≤1	1.8	3.6				
Tack-free time (min)		30	30	30	30	30	60	60	60	80	80	80	100	100	100	90	90	90				
Resistance to weather		◎	◎	◎	◎	◎	△	△	△	△	△	△	△	△	△	×	×	×				

As shown in Table X1, the coating materials of the present invention for automobiles exhibit excellent resistance to chipping, vibration, sprayed saline water and weather, and also very high curing speed, even when cured at low temperature and
5 for a short time, and also when it is thin.

<EXAMPLES Y Series>

PRODUCTION EXAMPLE Y1

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10 [Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
rubber]

15 The three-component copolymerization was effected continuously in a stainless steel polymerization reactor having an essential capacity of 100L, equipped with agitator blades (agitating rotation speed: 250rpm), wherein hexane, ethylene, propylene and 5-vinyl-2-norbornene were continuously supplied at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$ and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols per hour, respectively, also continuously.

20 The copolymerization effected under the above conditions produced the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution, continuously withdrawn from the reactor bottom, to terminate

the polymerization. The polymer was separated from the solvent by steam-stripping the solution, and dried at 55°C for 48 hours under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber thus produced contained ethylene at 68% by mol, and had an iodine value of 10, intrinsic viscosity $[\eta]$ of 0.2dl/g, and Mw/Mn of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber containing dimethoxymethylsilyl group.

EXAMPLE Y1

The silyl-containing copolymer rubber prepared in PRODUCTION EXAMPLE Y1 was incorporated with a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), limestone powder (Maruo Calcium, Snowlite SSTM) and salt cake ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), and the mixture was well kneaded by a 3-paint roll unit, to produce the major ingredient for the sealant. The respective amounts as the part by weight are given in Table Y1. Dibutyl tin bisacetylacetonate (NITTO KASEI, U-220TM) was used as the curing

catalyst.

The major ingredient and curing catalyst as the component of the test sample were well kneaded, formed into an around 1.5mm thick sheet and cured in an oven under the conditions of 23°C and 50%RH for 7 days and then 50°C and 70%RH for another 7 days. The composition is given in Table Y1.

The cured sheet sample was tested in accordance with JIS Z-0208 (the moisture-permeability test for moisture-proof packing materials) under the temperature/humidity conditions B (40°C and 90%RH).

The sample was evaluated by moisture permeability according to the following three grades:

○ : Permeability: 1 to 25g/m²·24hr

△ : Permeability: 25 to 50g/m²·24hr

× : Permeability: 50g/m²·24hr or more

The result is given in Table Y1.

REFERENCE EXAMPLE Y1

The sample was prepared and tested in the same manner as in EXAMPLE Y1, except that the silyl-containing copolymer rubber prepared in PRODUCTION EXAMPLE Y1 was replaced by the isobutene polymer with a group containing reactive silicon at both terminals, prepared by the method disclosed by Japanese Patent Laid-open Publication No. 209539/1999 (paragraphs 0044 to 0053). The moisture permeability result is given in Table Y1, wherein

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the isobutene polymer with a group containing reactive silicon
is listed in the column of the component (A).

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Table Y1

	EXAMPLE	REFERENCE EXAMPLE
	Y1	Y1
Compositions (Major ingredient) Component (A2) Silyl-containing copolymer rubber, prepared in PRODUCTION EXAMPLE 1 Isobutene polymer containing reactive silicon Other additives Process oil (PS-32) Limestone powder (Snowlight SS) $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ Component (H) Curing agent (U-220)	100 - 100 460 2 4	- 100 100 460 2 4
Characteristics of cured product Moisture permeability ($\text{g}/\text{m}^2 \cdot 24\text{hr}$) Thickness (mm)	0 1.637	0 1.868

EXAMPLES Y2 to Y5

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5 The major ingredient was prepared for each of EXAMPLES Y2 to Y5, where the silyl-containing copolymer rubber prepared in PRODUCTION EXAMPLE Y1 was well kneaded by a 3-roll unit together with the additives: aging inhibitors (Ciba-Geigy Japan, Irganox 1010; Sumitomo Chemical, Sumisorb 400TM, and Sankyo, Sanol LS-765TM); light stabilizers (Sanshin Kagaku Kogyo, Sandant NBCTM; and ACC, CYASORB UV-1084TM); light-curable resin (TOAGOSEI, Aronix M-400TM); thixotropy imparting agent (Kusumoto Kasei, Disparlon #305TM); silane coupling agents (Nippon Unicar, A-1310 and A-187TM). Each composition is given in Table Y2.

15 The curing agent was prepared by the following procedure: a mixture comprising a curing catalyst (Sankyo Organic Chemicals, Ltd., SCAT-27TM) and other components was manually kneaded in a disposal cup and stirred 3 times at 10,000 rpm each for 10 minutes by a homogenizer (Nihon Seiki Sesakusho Co., Ltd., Excel Auto Homogenizer). Each composition is given in Table Y2.

20 The test piece was prepared in accordance with JIS A-5758/1992 specifying the method of preparing the test piece for tensile adhesion test; the composition comprising the major ingredient and curing agent (composition is given in Table Y2) was put in the H-shape frame of glass or aluminum substrate after it was sufficiently kneaded, and cured in an oven under the conditions of 23°C and 50%RH for 7 days and 50°C and 70%RH for another 7

days.

Three types of materials were used to prepare substrates for the H-type tensile test; float glass (Koen-sha, designated by Japan Sealant Industry Association, 3 by 5 by 0.5cm in size) in accordance with JIS A-5758/1992, pure aluminum (Taiyu Kizai, A1100P, 5 by 5 by 0.2cm in size) in accordance with JIS H-4000, and heat ray reflective glass (KLSTM, 5 by 5 by 0.6cm in size) coated with thermally fused TiOx.

Each of these H-shapes was washed with methylethylketone (Wako-Junyaku, special grade) and wiped with clean cotton cloth, before it was filled with the composition.

The H-shape test piece thus prepared was tested by the method of testing tensile adhesion in accordance with JIS A-5758/1992, wherein it was stretched at a tensile speed of 50mm/minute in a constant-temperature chamber kept at 23°C and 65±5%. The cohesion fracture (CF)/thin-coat fracture (TCF)/adhesion fracture (AF) ratio shown in Table Y3 was determined by visual observation of the cross-sections of the tensile-tested pieces.

As shown in Table Y3, all of the compositions prepared in EXAMPLES Y2 to Y5 exhibit good adhesion to the substrate.

REFERENCE EXAMPLES Y2 to Y5

The composition was prepared for each of REFERENCE EXAMPLES Y2 to Y5 and tested in the same manner as in EXAMPLES Y2 to Y5, except that the silyl-containing copolymer rubber prepared in

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PRODUCTION EXAMPLE Y1 was replaced by the isobutene polymer prepared in REFERENCE EXAMPLE Y1 to have a group containing reactive silicon at both terminals. The test results are given in Table Y4.

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Table Y2

EXAMPLES					
	Y2	Y3	Y4	Y5	
[Major ingredients]					
Polymer prepared in PRODUCTION EXAMPLE Y1					
PS32	100	100	100	100	100
EDS-D10A	112.5	135	157	180	180
PO320B10	62.5	75	87.5	100	100
Talc LMR	22.5	270	315	360	360
Sandant NBC	12.5	150	175	200	200
Aronix M400	3	3	3	3	3
Disparlon #306	3	3	3	3	3
Sumisorp 400	5	5	5	5	5
Sanol LS786	1	1	1	1	1
Irganox 1010	1	1	1	1	1
A-187	1	1	1	1	1
A1310	2	2	2	2	2
	4	4	4	4	4
[Curing agents]					
SCAT-27	4	4	4	4	4
PS32	12.5	15	17.5	2.0	2.0
CB#30	2.5	2.5	2.5	2.5	2.5
Na2SO4• 10H2O	4	4	4	4	4
Snowlite SS	25	30	35	40	40
Polymer content (%)	14.4	12.4	10.9	9.7	9.7

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Table Y3

No.	Substrates	50% Tensile stress M50 (kgf/cm ²)	Max.tensile stress Tmax (kgf/cm ²)	Elongation at the max. load Emax (%)	Fractured conditions (%)		
					CF	TCF	AF
EXAMPLE Y2	Float glass	1	4.46	6.17	87	0	0
		2	4.66	6.20	86	0	0
		average	4.64	6.19	87	0	0
	Pure aluminum	1	4.39	6.21	91	1	0
		2	4.37	6.10	87	0	0
		average	4.38	6.16	98	0	0
EXAMPLE Y3	KLS	1	4.67	6.22	88	1	0
		2	4.69	6.25	93	0	0
		average	4.68	6.24	91	0	0
	Float glass	1	4.69	6.26	86	2	0
		2	4.71	6.27	84	2	0
		average	4.70	6.27	85	2	0
EXAMPLE Y4	Pure aluminum	1	4.37	6.18	87	1	0
		2	4.35	6.12	82	0	0
		average	4.36	6.15	85	0	0
	KLS	1	4.80	6.32	80	3	0
		2	4.83	6.36	79	3	0
		average	4.82	6.34	80	3	0
EXAMPLE Y5	Float glass	1	4.66	6.11	72	2	0
		2	4.69	6.02	74	0	0
		average	4.68	6.07	73	1	0
	Pure aluminum	1	4.36	6.11	79	7	0
		2	4.33	6.08	77	3	0
		average	4.35	6.05	78	5	0
EXAMPLE Y6	KLS	1	4.67	6.13	75	1	0
		2	4.70	6.11	72	1	0
		average	4.69	6.12	74	1	0
	Float glass	1	4.33	6.27	70	2	0
		2	4.30	6.23	73	1	0
		average	4.32	6.25	72	1	0
EXAMPLE Y7	Pure aluminum	1	3.76	5.89	73	2	0
		2	3.72	5.91	77	2	0
		average	3.74	5.90	75	2	0
	KLS	1	4.32	6.18	78	0	0
		2	4.37	6.11	71	0	0
		average	4.35	6.15	75	0	0

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Table Y4

No.	Substrates		50% Tensile stress M50 (kgf/cm ²)	Max tensile stress T _{max} (kgf/cm ²)	Elongation at the max. load E _{max} (%)	Fractured conditions (%)		
						CF	TCF	AF
REFERENCE EXAMPLE Y2	Float glass	1	4.67	6.98	98	100	0	0
		2	4.76	6.87	91	100	0	0
		average	4.72	6.93	95	100	0	0
	Pure aluminum	1	4.43	6.51	91	100	0	0
		2	4.38	7.01	106	99	1	0
		average	4.41	6.76	99	100	1	0
REFERENCE EXAMPLE Y3	KLS	1	4.79	7.25	99	98	2	0
		2	4.66	7.43	105	100	0	0
		average	4.73	7.34	102	99	1	0
	Float glass	1	4.74	6.70	89	95	5	0
		2	4.79	7.10	93	85	15	0
		average	4.77	6.90	91	90	10	0
REFERENCE EXAMPLE Y4	Pure aluminum	1	4.38	6.81	94	100	0	0
		2	4.43	6.53	85	95	6	0
		average	4.41	6.67	90	98	3	0
	KLS	1	4.92	6.58	81	90	10	0
		2	5.03	6.86	83	90	10	0
		average	4.98	6.72	82	90	10	0
REFERENCE EXAMPLE Y5	Float glass	1	4.75	6.32	77	98	2	0
		2	4.79	6.43	77	100	0	0
		average	4.77	6.38	77	95	1	0
	Pure aluminum	1	4.38	6.34	86	85	16	0
		2	4.53	5.94	75	100	0	0
		average	4.46	6.14	81	100	8	0
REFERENCE EXAMPLE Y5	KLS	1	4.68	6.29	81	99	1	0
		2	4.83	6.17	74	90	10	0
		average	4.76	6.23	78	100	6	0
	Float glass	1	4.28	6.00	79	95	5	0
		2	4.36	5.98	76	100	0	0
		average	4.32	5.99	78	98	3	0
REFERENCE EXAMPLE Y5	Pure aluminum	1	3.92	5.38	79	95	5	0
		2	3.99	6.13	87	98	2	0
		average	3.96	5.76	83	97	4	0
	KLS	1	4.37	6.35	85	100	0	0
		2	4.83	5.86	70	100	0	0
		average	4.60	6.11	78	100	0	0

The curing speed and weather resistance tests were conducted for the compositions prepared in EXAMPLES Y1 to Y5 and REFERENCE EXAMPLES Y1 to Y5. The results are given in Table Y5.

5 The curing speed and resistance to weather were measured for the compositions prepared in EXAMPLES Y1 to Y5 and REFERENCE EXAMPLES Y1 to Y5 by the following methods.

1) Curing speed

10 Each of the compositions comprising the major ingredient and catalyst was measured for curing speed (film expandability) at room temperature by the method in which the composition was cured at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and then released from the mold. Thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm.

15 <Evaluation standards for curing speed>

× : The cured portion was less than 0.5mm thick

△ : The cured portion was 0.5mm thick or more but less than 1mm

○ : The cured portion was 1mm thick or more.

20 2) Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120

minutes/rainfall for 18 minutes

Black panel temperature: $63 \pm 2^{\circ}\text{C}$

Tank inside temperature: $40 \pm 2^{\circ}\text{C}$

Total light irradiation time: 500 hours

- 5 The tested test piece was visually observed, to evaluate its resistance to weather according to the following three grades:

○ : No cracks or molten portion observed

△ : Cracks or molten portion observed slightly

× : Cracks or molten portion observed

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Table Y5

	Resistance to weather	Curing speed
EXAMPLE Y1	○	○
EXAMPLE Y2	○	○
EXAMPLE Y3	○	○
EXAMPLE Y4	○	○
EXAMPLE Y5	○	○
REFERENCE EXAMPLE Y1	△	×
REFERENCE EXAMPLE Y2	△	×
REFERENCE EXAMPLE Y3	△	×
REFERENCE EXAMPLE Y4	△	×
REFERENCE EXAMPLE Y5	△	×

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<EXAMPLES Z Series>

PRODUCTION EXAMPLE Z1

[Production of silyl-containing
ethylene/propylene/5-vinyl-2-norbornene random copolymer
5 rubber]

The three-component copolymerization was effected
continuously in a stainless steel polymerization reactor having
an essential capacity of 100L, equipped with agitator blades
(agitating rotation speed: 250rpm), wherein hexane, ethylene,
10 propylene and 5-vinyl-2-norbornene were continuously supplied
at 60L, 2.5kg, 4.0kg and 380g per hour, respectively, from the
reactor side into the liquid phase, and hydrogen, $\text{VO}(\text{OC}_2\text{H}_5)_2\text{Cl}$
and $\text{Al}(\text{Et})_{1.5}\text{Cl}_{1.5}$ as the catalysts at 700L, 45mmols and 315mmols
per hour, respectively, also continuously.

15 The copolymerization effected under the above conditions
produced the ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber in a form of uniform solution.

A small quantity of methanol was added to the polymer solution,
continuously withdrawn from the reactor bottom, to terminate
20 the polymerization. The polymer was separated from the solvent
by steam-stripping the solution, and dried at 55°C for 48 hours
under a vacuum.

The ethylene/propylene/5-vinyl-2-norbornene random
copolymer rubber thus produced contained ethylene at 68% by mol,

and had an iodine value of 10, intrinsic viscosity $[\eta]$ of 0.2dl/g, and M_w/M_n of 15.

Two% toluene solution (0.3g) of chloroplatinic acid and 1.5g of methyldimethoxysilane were added to 100g of the ethylene/propylene/5-vinyl-2-norbornene random copolymer rubber, and they were allowed to react with each other at 120°C for 2 hours. The excess methyldimethoxysilane and the solvent were distilled off from the effluent. This produced 101.5g of the copolymer rubber containing dimethoxymethylsilyl group.

10 EXAMPLES Z1 to Z3, and COMPARATIVE EXAMPLE Z1

20 The polymer prepared in PRODUCTION EXAMPLE Z1, 100 parts was incorporated with 30 parts of a paraffin-based process oil (Idemitsu Kosan, Diana Process Oil PS-32TM), 130 parts of a butyl-based hot melt (Yokohama Rubber, Hamatite HOTMELT M-120), 6 parts of salt cake (reagent, $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), 3 parts of tin octylate (NITTO KASEI, Neostann U-28TM) and 0.75 part of lauryl amine (Wako Jun-yaku), all parts by weight. The mixture was well kneaded by a 3-paint roll unit, to produce the test sample for EXAMPLE Z1.

20 The compositions were prepared for EXAMPLES Z2 and Z3 in the same manner as in EXAMPLE Z1, except that contents of the butyl-based hot melt were changed to 303.3 and 1169 parts by weight, respectively, to prepare the test samples. The composition was also prepared for COMPARATIVE EXAMPLE Z1 in the

same manner as in EXAMPLE Z1, except that the polymer was incorporated only with the hot melt to prepare the test sample. These samples were used for the tensile test.

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10 The test sample was prepared in accordance with JISA-6850/1976 which specifies the method of sample preparation for testing tensile adhesion strength of adhesives. Each composition was spread over and pressed to an aluminum plate (Taiyu Kizai, A-1050P, 2.5 by 10 by 0.3mm in size, specified by JIS H-4000) as the substrate, which was washed with methylethylketone (Wako Jun-yaku) and wiped with clean cotton cloth beforehand. Each composition was cured as the sealing material at 50°C for 4 days in an oven.

15 The test sample prepared by the above method was stretched at 50mm/minute in a constant-temperature chamber kept at 23°C and RH65±5%, in accordance with JIS A-6850 for the tensile adhesion testing method. The results are given in Table Z1.

REFERENCE EXAMPLES Z1 to Z3

20 The isobutene polymer was prepared and tested in each of REFERENCE EXAMPLES Z1 to Z3 in the same manner as in corresponding EXAMPLE Z1, Z2 or Z3, except that the silyl-containing copolymer rubber prepared in PRODUCTION EXAMPLE Z1 was replaced by the isobutene polymer with a group containing reactive silicon at both terminals, prepared according to the method disclosed by Japanese Patent Laid-open Publication No.209540/1999

(paragraphs 0041 to 0050). The results are given in Table Z1, together with the results of the curing speed and weather resistance tests.

The curing speed and resistance to weather were measured by the following methods.

1) Curing speed

Each of the compositions comprising the major ingredient and catalyst as described above was measured for curing speed (film expandability) at room temperature by the following method:

The composition was cured in a chamber kept at 23°C and 50%RH for 24 hours in a mold (20 by 80 by 5mm in size), and then released from the mold. Thickness of the cured portion was measured by a dial gauge of weak spring force to 0.1mm.

<Evaluation standards for curing speed>

× : The cured portion was less than 1mm thick.

○ : The cured portion was 1mm thick or more.

2) Weather resistance test

The accelerated weather resistance test was conducted in accordance with JIS B-7753 under the following conditions:

Analyzer : Sunshine Carbon Arc weatherometer

Light irradiation/rainfall cycles: Irradiation for 120 minutes/rainfall for 18 minutes

Black panel temperature: 63±2°C

Tank inside temperature: 40±2°C

Total light irradiation time: 500 hours

The tested test piece was visually observed, to evaluate its resistance to weather according to the following three grades:

- : No cracks or molten portion observed
- 5 △ : Cracks or molten portion observed slightly .
- × : Cracks or molten portion observed

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Table Z1

		Max. tensile stress T _{max} (kgf/cm ²)	Elongation at the max. load E _{max} (%)	Curing speed	Resistance to weather
EXAMPLE Z1	1	2.1	90	○	○
	2	1.9	100		
	average	2.0	95		
EXAMPLE Z2	1	2.0	76	○	○
	2	1.8	84		
	average	1.9	80		
EXAMPLE Z3	1	1.8	55	-	-
	2	1.6	65		
	average	1.7	60		
COMPARATIVE EXAMPLE Z1	1	1.2	45	×	△
	2	1.4	35		
	average	1.3	40		
REFERENCE EXAMPLE Z1	1	2.1	32	×	△
	2	2.1	28		
	average	2.1	30		
REFERENCE EXAMPLE Z2	1	1.5	12	×	△
	2	1.7	8		
	average	1.6	10		
REFERENCE EXAMPLE Z3	1	1.6	18	-	-
	2	1.6	22		
	average	1.6	20		

EXAMPLE Z4 and REFERENCE EXAMPLE Z4

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5 The cured sheet of about 2mm thickness was prepared using
the composition produced in each of EXMPLE Z1 and REFERENCE
EXAMPLE Z1. The cured sheet was stamped out into the No.3
dumbbell-shaped test piece in accordance with JIS K-6301. The
results of tensile test are shown in Table Z2 (EXAMPLE Z4) and
Table Z3 (REFERENCE EXAMPLE Z4), respectively. The test piece
was cured at 23°C for 7 days and at 50°C for another 7 days,
and then taken out for measurement of H-type mechanical
10 characteristics.

The test was conducted at a stretching speed of 200 mm/min
in a thermostat kept at 23, 50 and 70°C according to the method
of the tensile test specified by JIS K-6301.

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Table Z2

EXAMPLE		50% Tensile stress M50 (kgf/cm ²)	100% Tensile stress M100 (kgf/cm ²)	Max. tensile stress T _{max} (kgf/cm ²)	Elongation at the max. load E _{max} (%)
Z4	1	1.1	1.6	10.0	560
	2	0.9	1.5	10.5	480
	3	1.0	1.7	9.5	520
	Average	1.0	1.6	10.0	520
50 (°C)	1	0.9	1.5	4.5	390
	2	0.7	1.5	5.5	370
	3	0.8	1.2	5.0	380
	Average	0.8	1.4	5.0	380
70 (°C)	1	0.8	1.5	4.1	340
	2	0.8	1.5	4.0	340
	3	0.7	1.4	3.9	320
	Average	0.8	1.5	4.0	330

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Table Z3

REFERENCE EXAMPLE Z4	50% Tensile stress M50 (kgf/cm ²)	100% Tensile stress M100 (kgf/cm ²)	Max. tensile stress T _{max} (kgf/cm ²)	Elongation at the max. load E _{max} (%)
23 (°C)	1	1.0	1.7	9.2
	2	1.1	1.7	11.0
	3	1.2	1.9	11.7
	Average	1.1	1.8	10.6
50 (°C)	1	0.9	1.5	4.4
	2	0.8	1.5	5.1
	3	1.0	1.7	5.8
	Average	0.9	1.5	5.1
70 (°C)	1	1.0	1.7	5.0
	2	0.9	1.6	4.4
	3	0.9	1.5	3.5
	Average	0.9	1.6	4.3